Environmental Impacts Assessment of Recycling of Construction and Demolition Waste

Butera, Stefania

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Stefania Butera

DTU Environment
Department of Environmental Engineering

PhD Thesis
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Address: DTU Environment
Department of Environmental Engineering
Technical University of Denmark
Miljoevej, building 113
2800 Kgs. Lyngby
Denmark

Phone reception: +45 4525 1600
Fax: +45 4593 2850

Homepage: http://www.env.dtu.dk
E-mail: reception@env.dtu.dk

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Preface

This thesis is organised in two parts: the first part puts into context the findings of the PhD in an introductive review, while the second part consists of the papers listed below. These will be referred to in the text by their paper number, written in Roman numerals **I-IV**.

I **Butera S**, Christensen TH, Astrup TF. (2014). Composition and leaching of construction and demolition waste: inorganic elements and organic compounds. *Journal of Hazardous Materials*, 276, 302–311. doi:10.1016/j.jhazmat.2014.05.033

II **Butera S**, Hyks J, Christensen TH, Astrup TF. Construction and demolition waste: comparison of standard up-flow column and down-flow lysimeter leaching tests. *Submitted, 2014.*

III **Butera S**, Trapp S, Astrup TF, Christensen TH. Soil retention of hexavalent chromium released from construction and demolition waste in a road-base application scenario. *Submitted, 2014.*

IV **Butera S**, Christensen TH, Astrup TF. Life Cycle Assessment of construction and demolition waste management. *Submitted, 2014.*

In this online version of the thesis, the papers detailed above are not included but can be obtained from electronic article databases, e.g. via www.orbit.dtu.dk or on request from DTU Environment, Technical University of Denmark, Miljøvej, Building 113, 2800 Kgs. Lyngby,Denmark, reception@env.dtu.dk.

In addition, six conference proceedings and the following publication, not included in this thesis, were also concluded as part of this PhD study:

Allegrini E, **Butera S**, Kosson DS, van Zomeren A, van der Sloot HA, Astrup, TF. (2014). Life cycle assessment and residues leaching: importance of parameter, scenario and leaching data selection. Submitted to *Waste Management*.

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Alberto, *Trine* and *Valentina* are kindly acknowledged for their support with sampling (and boring – and occasionally overnight) laboratory activities. Thanks to *Anders* for saving me from Easetech many times, and for his help with the Dansk sammenfatning. I would like to express my gratitude to *Sinh Hy Nguyen* and *Susanne Kruse* for their patient and careful analytical work. Thanks also to *Torben Dolin* and *Lisbet Brusendorff* for their invaluable help with the figures, and to *Anne Harsting* for taking good care of PhDs at the Department and for her constant smile.

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Thanks to *my friends* here in Denmark (or wherever life has brought them) and in Italy for loving me despite my evident sleep-deprivation problem – just to mention one. Also for making good food and better music. A special thanks to *P*, for making my day almost every day.

Finally, thanks to *Mr. G.* for not even pretending to be interested in "the story of Cr(VI)" but for listening anyway. And for way more, clearly.

And obviously, I would like to thank *my parents* and *my brothers* for their patience in dealing with me for so many years and for their love and support.

Summary

Construction and demolition waste (C&DW) is waste derived from the construction, demolition and renovation of buildings and civil infrastructure. With 900 million tons generated every year in Europe, it is the largest waste stream on the continent. C&DW is mainly constituted of mineral fractions, i.e. soil and stones, concrete, asphalt and masonry, and as such it has the potential to be used as aggregate in the construction sector. A typical application is in an unbound state as filler in road structures. This practice offers evident benefits in terms of resource savings, however it might lead to potential adverse impacts, especially related to the water-borne emission of pollutants, which need to be avoided. This requires first of all an estimation of their magnitude, and so the goal of this PhD is to provide an assessment of potential environmental impacts related to C&DW utilisation.

C&DW is characterised by significant variability, especially in terms of leaching. Different levels of Ca, Ba, Cl^-, Cr, K, Li, Mg, Na, Sr, Se, Si, SO_4 and V are in evidence, depending on the ageing level (and therefore on the extent of carbonation) as well as the content of masonry. Both aspects may be optimised by appropriate measures within the C&DW waste management system, for instance by promoting source segregation of the concrete fraction stream or actively pursuing its carbonation. We found that leaching of Se, and to a lower extent Cr, Sb, SO_4, Cl^-, appears critical for C&DW in relation to existing national and European regulations. Cr mainly exists in C&DW leachates as anionic species, which may be interpreted as hexavalent species (mainly chromate). Despite being banned several decades ago, PCBs are still found in C&DW and in concrete raw materials, albeit in low, non-critical concentrations. This highlights their ubiquitous environmental presence.

Several methods may be used to investigate leaching from granular C&DW, one of which is percolation tests. Compared to down-flow lysimeters with uncrushed C&DW, this study found that the use of standard up-flow columns, with materials below 4mm in particle size, may introduce differences especially in terms of pH, which in turn may affect the leaching of Al, As, Ba, Cu, DOC, Mg, Mn, P, Pb, Sb, Se, Si and Zn as a consequence of the crushing process, which results in the exposure of fresh, un-carbonated surfaces. However, when the scope involves quantifying cumulative release, standard up-flow columns may be considered appropriate, while for estimating early concentrations, relying on standard up-flow columns may be more problematic (e.g. Al, As, Cu, DOC, Mg, Mn, P, Pb, Sb, Se, Si and Zn), and the relation-

ship between testing conditions and field conditions should be evaluated critically.

Owing to its high toxicity and significant mobility, especially at high pH levels, Cr(VI) is one of the elements of concern found in C&DW leachates. Its fate in the sub-soil below road applications was assessed experimentally, and its vertical migration was then predicted through a model. Interactions with sub-soil particles, namely reduction to immobile Cr(III), are responsible for the retention of Cr(VI) in the first 70 cm of sub-soil below the C&DW sub-base. Temperate climates might inhibit the already slow reduction kinetics, resulting in Cr(VI) migration up to 2 m. The same case applies in situations characterised by high infiltration rates, such as unpaved roads, cracked asphalt cover or heavy rain events.

By using holistic tools such as life cycle assessment (LCA) a general evaluation of the environmental consequences of C&DW utilisation system was provided. Although for most impact categories C&DW utilisation in road sub-bases does not provide environmental savings in absolute terms, it is generally less hazardous than when being landfilled (excluding toxicity impacts). On the other hand, landfilling appears better than C&DW utilisation when considering toxicity categories, owing to lower leaching in landfill scenarios over a 100-year time horizon. Oxyanions play a predominant role in leaching impacts, rather than cationic metals, and accurate modelling of Cr(VI) fate is essential to the results, while the heterogeneity of C&DW leachates does not play a crucial role in LCA results. C&DW carbonation leads to a trade-off between reducing global warming impacts and increasing toxic impacts related to the higher leaching of oxyanions. While leaching appears as the major problem relating to C&DW utilisation in LCA terms, uncertainties related to methodological aspects of leaching modelling in LCA should be acknowledged.

Dansk sammenfatning

Bygge- og anlægsaffald (B&A affald) er affald, der genereres i forbindelse med konstruktion, nedrivning og renovering af bygninger og civil infrastruktur. Med 900 millioner ton genereret i Europa hvert år, repræsenterer B&A affald den største enkeltstående affaldsstrøm i Europa. B&A affald består hovedsagelig af mineralske fraktioner, dvs. jord og sten, beton, asfalt, tegl og mursten, og har derfor potentiale til at blive anvendt som fyldmateriale i byggesektoren. Typisk erstatter B&A affald i ubunden form stabilgrus, som ellers ville være anvendt f.eks. i vejbygning. Selv om denne praksis medfører ressourcebesparelser i samfundet, bør potentielle negative påvirkninger undgås - især i relation til udvaskning af problematiske stoffer. Dette kræver en systematisk vurdering af udvaskningen fra affaldet og de derved afledte miljøpåvirkninger. Formålet med denne Ph.d. er at give en vurdering af de potentielle miljøeffekter i et livscyklusperspektiv ved anvendelsen af B&A affald.

B&A affald er et heterogent materiale og udvaskningen af potentielt problematiske stoffer varierer. Forskellige niveauer af Ca, Ba, Cl^-, Cr, K, Li, Mg, Na, Sr, Se, Si, SO_4, og V blev observeret, afhængig af materialets karbonatisering, samt mængden af tegl og mursten. Begge dele kan optimeres i relation til håndtering og behandling af B&A affald, for eksempel ved at øget kildesortering og dermed øget renhed af betonfraktionen, eller aktivt at tilstræbe karbonatisering af betonen. I sammenligning med eksisterende nationale og europæiske grænseværdier, blev udvaskningen af selen, og i mindre omfang krom, antimon, sulfat, og klorid, fastlagt som værende kritisk for udnyttelsen af B&A affald. Krom findes i perkolatet fra B&A affald hovedsageligt i anionisk form, dvs. primært som hexavalent krom (Cr(VI)). Selvom PCB blev forbudt for flere årtier siden, findes forbindelserne stadig i B&A affald til genanvendelse; dog i lave, ikke-kritiske koncentrationer.

Adskillige fremgangsmåder blev anvendt til at undersøge udvaskningen fra knust B&A affald, bl.a. "up-flow" kolonneforsøg, som under standardiserede forhold simulerer infiltrationen af vand gennem nedknust materiale. Forholdene i de standardiserede kolonneforsøg blev sammenlignet med "down-flow lysimeterforsøg", hvor B&A affald ikke blev nedknust og hvor vandtilsætning og - gennemstrømning i højere grad simulerede forholdene i byggeprojekter. Der blev observeret forskelle i udvaskningen, især med hensyn til pH, hvilket kan påvirke udvaskningen af Al, As, Ba, Cu, DOC, Mg, Mn, P, Pb, Sb, Se, Si og Zn. pH-forskellene kan være en konsekvens af knusningen af materialet i standardtesten, hvilket resulterer i eksponering af friske, ikke-

karbonatiserede overflader. På trods af forskelle i udvaskningen, særligt perkolatkoncentrationer ved lave væske-faststof forhold (L/S), giver de standardiserede upflow kolonner dog for de fleste elementer et rimeligt billede af den kumulative udvaskning fra materialerne. Forskelle mellem testbetingelser og betingelser for udvaskning i fuldskala bør dog vurderes kritisk ved vurdering af konkrete situationer.

På grund af høj toksicitet og betydelig mobilitet, især ved høj pH, er Cr(VI) et af de mest kritiske elementer i B&A affald perkolat. Cr(VI) mobilitet i de nedre jordlag i forbindelse med anvendelse som stabilgrus ved vejbyggeri blev vurderet eksperimentelt, og den vertikale migration blev modelleret. Interaktioner med partikler i undergrunden, primært i form af reduktion til immobil og ikke-toksisk Cr(III), er den primære mekanisme for begrænsning af udvaskningen af Cr(VI) fra de første 70 cm af jord under et B&A affald lag. Kinetikken for reduktion af Cr(VI) til Cr(III) er temperaturafhængig og vil i tempererede områder potentielt kunne medføre en øget Cr(VI) migration på op til 2 m under B&A affald laget. Tilsvarende øget migration kan desuden ske i situationer karakteriseret ved høje infiltrationsrater, såsom ikke-asfalterede veje, revnet asfaltdækning, eller i forbindelse med skybrud.

Ved brug af holistiske vurderingsværktøjer, såsom livscyklusvurdering (LCA), blev en generel evaluering af de samlede miljømæssige konsekvenser ved udnyttelse af B&A affald fastlagt. Selvom udnyttelsen af B&A affald til vejbygning ikke i sig selv medfører miljømæssige besparelser, så er miljøpåvirkningerne generelt lavere end alternativ deponering (dog undtaget toksiske effekter). Transport af B&A affald påvirker resultatet betydeligt i relation til f.eks. klimapåvirkningen. I forbindelse med de toksiske kategorier, bidrager scenarier med deponering med lavere påvirkninger som følge af en lavere netto udvaskning i deponeringsanlæggene. Oxyanioner spiller i højere grad end metaller en fremtrædende rolle i påvirkninger fra udvaskning. En specifik modellering af Cr(VI) emissionerne er derfor afgørende for de overordnede resultater. Typiske variationer i udvaskning fra B&A affald spiller ikke en afgørende rolle i forhold til LCA resultaterne. Karbonatisering af B&A affald medfører potentielt en reduktion af klimapåvirkningerne, men også øgede toksiske påvirkninger som følge af forhold til øget udvaskning. Udvaskningen fra B&A affald fremstår samlet set som et kritisk aspekt i relation til LCA resultaterne. Der bør derved sættes fokus på de metodiske aspekter omkring inkludering af udvaskning i LCA modellering, ligesom usikkerhederne forbundet med dette bør anerkendes.

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1 Introduction

1.1 Background

The utilisation of solid residues, e.g. construction and demolition waste (C&DW) and municipal solid waste incineration (MSWI) bottom ash (BA), is an increasingly common option for solid residues management. Their potential for utilisation as aggregate derives first of all from their plentiful availability (see Table 1), their physical properties, similar to those of common natural aggregates, and from their relatively low contaminant content. While utilisation in bound applications (e.g. in concrete) still presents some technical drawbacks, owing to the reduced mechanical properties of residues compared to natural aggregates (Silva et al., 2014), one of the most widespread options is application in an unbound form as a filler material in road bases, sub-bases, embankments and other earthworks (Hendriks and Janssen, 2001; Tam and Tam, 2006). Utilisation rates in Denmark are 96% and 99.7% for C&DW and MSWI BA, respectively (Danish EPA, 2011). These residues are prevalently inorganic and incombustible, and therefore landfilling represents the only alternative, although not preferred, management option.

Table 1 Yearly generation (in $Tg \cdot y^{-1}$, corresponding to thousands $t \cdot y^{-1}$) of total solid waste, C&DW and BA from incineration of waste in Europe and Denmark. Percentages in parentheses represent the fraction compared to the total waste generated.

[$Tg \cdot y^{-1}$]	EU-27 (2008)[a]	Denmark (2011)[b]
Total Waste generated	2,600,000	9,100
C&DW	860,000 (33%)	2,400 (26%)
Incineration BA	35,000 (1%)	700 (8%)

[a] Source: Eurostat (Schör, 2011)

[b] Source: (Danish EPA, 2013)**;** data for BA were estimated from the total amount of waste incinerated.

C&DW represents waste resulting from the construction, renovation and demolition of buildings and civil infrastructures. Its composition, given in Table 2 for the case of Denmark, indicates that mineral fractions – soil and stone, concrete, asphalt and masonry – constitute the largest portion. However, other fractions are also present, typically metal, wood, glass and plastic, depending on the building practices and materials used in the area. In this thesis, nonetheless, the term C&DW is used primarily to indicate the mineral fraction of the entire waste stream, i.e. soil and stones, concrete, asphalt and masonry.

C&DW represents the largest waste stream in Europe (Schör, 2011); hence, it has high importance. Specific targets concerning its recycling have been included in the Waste Framework Directive (2008/98/EC) (European Commission, 2008), and substantial work is carried out at European level regarding its management (e.g., EU Commission, 2011; Monier et al., 2011), and end-of-waste criteria (Saveyn et al., 2014). The main goal is to minimise its disposal and emphasise its potential as a resource, at the same time guaranteeing a proper level of environmental protection, as there are in fact a number of potential harmful compounds in C&DW: organic pollutants, such as polycyclic aromatic hydrocarbons (PAHs) and polychlorinated biphenyls (PCBs), and several inorganic elements. PAHs might originate from contact with tar-containing fractions (e.g., asphalt) or from atmospheric deposition, while PCBs may derive from contamination from PCB-containing products such as sealing materials and paint. Furthermore, inorganic contaminants might derive from the use of alternative fuels (e.g., waste) during clinker production, from additives in cement or concrete production, e.g. coal fly ash (FA), or even be present naturally in raw materials. When C&DW is used in unbound road construction applications, the main environmental issue is represented by the leaching of harmful inorganic compounds and the subsequent potential contamination of underlying soil and groundwater.

Table 2 Composition of Danish C&DW in 2009 (Danish EPA, 2011).

	Fraction [%]
Soil and stone[a]	28.0
Concrete	25.8
Asphalt	19.1
Other C&DW	12.5
Not combustible	1.8
Others	6.9
Tiles	4.1
Other recyclables	1.8

[a] Depending on the country, soil and stone may sometimes be excluded from C&DW inventories.

1.2 Research objectives

The overall objective of this PhD project is to provide a sound assessment of potential environmental impacts related to C&DW utilisation. Specific goals are to:

- Characterise Danish C&DW in terms of elemental composition, PAH and PCB content and basic leaching behaviour;

- Assess the importance of using leaching data from different percolation leaching tests in environmental assessment of C&DW utilisation in road construction;
- Estimate releases from C&DW in percolation scenarios, and identify critical parameters with respect to existing limit values;
- Investigate the chemical speciation of chromium in C&DW leachate, and evaluate its interactions with the sub-soil and its overall mobility in relation to C&DW utilisation;
- Based on the information collected, develop life cycle inventory (LCI) data for C&DW utilisation and perform an environmental impact assessment, taking into account the effects of carbonation on leaching behaviour as well as its significance as a CO_2 sink and the fate of emitted pollutants in sub-soil.

1.3 Rationale and approach

A considerable number of studies exist regarding levels and mechanisms of contaminant release from cementitious materials; however, while most of them focus on laboratory-formulated cement-based materials (e.g., Garrabrants et al., 2002; Hillier et al., 1999; Kayhanian et al., 2009; López Meza et al., 2008; Mulugeta et al., 2009; Sanchez et al., 2003, 2002; van der Sloot, 2000; van der Sloot et al., 2008b) or reference materials (e.g., Delay et al., 2007; Kalbe et al., 2008, 2007; Krüger et al., 2012), little information describes leaching from *actual* C&DW intended for recycling (Engelsen et al., 2009, 2003; Galvín et al., 2012; López Meza et al., 2010, 2009, 2008; Mulugeta et al., 2011; Nielsen et al., 2006; van der Sloot, 2002, 2000; Wahlström et al., 2000). Despite a good deal of concern, the presence of organic pollutants such as PCBs and PAHs has rarely been evaluated (Kalbe et al., 2008; Krüger et al., 2012; Nielsen et al., 2006; Strufe et al., 2006; Wahlström et al., 2000). Therefore, the first step in the research was to collect a large number of C&DW samples from treatment facilities, for the basic and representative characterisation of Danish C&DW. **Section 2** contains a brief description of the materials, methods and tools used during this research. After analysing their elemental composition, the presence of organic pollutants of concern (PAHs and PCBs) was evaluated. Results from relatively simple batch tests allowed for identifying a smaller set of samples with different leaching behaviours, on which a more detailed set of leaching tests

was then performed. These results are described in detail in **Paper I**, and the main points are summarised in **Section 3** of this synopsis.

Over the years, extensive research has been carried out concerning methods for quantifying leaching from granular solid residues (e.g., Kalbe et al., 2008; Kosson et al., 2014, 1996; van der Sloot et al., 2008a). However, some key aspects still remain unsolved with respect to release estimations from C&DW in percolation scenarios, specifically the effects of particle size and flow conditions. Therefore, a comparison between standard up-flow saturated columns on size-reduced material and lysimeters with downwards intermittent flow on coarse-grained C&DW was carried out on five of the collected samples (selected based on the abovementioned batch tests). This also allowed the identification of inorganic constituents of concern with respect to leaching, while an investigation into the mechanisms behind the observed differences was carried out based on pH static experiments. Results of this part of the work are reported in **Paper II** and **Section 3** of this thesis.

Based on the outcomes of the leaching tests, chromium was identified as one of the elements of concern. Once released into soil from a road sub-base application, chromium's oxidation state – and thus its toxicity – depends on its interactions with the soil. Therefore, the focus was shifted to Cr(VI) fate in sub-soil after being released from C&DW. Many authors have focused on Cr(VI) reduction in acidic environments and in relation to soils with high organic matter (e.g., Fonseca et al., 2009; Jardine et al., 1999; Kumpiene et al., 2006); however C&DW reuse scenarios are typically characterised by organic matter-poor sub-soils in contact with highly alkaline leachates containing potentially adsorption-competing anions. To address these issues, experiments were coupled with a transport model to estimate Cr(VI)'s overall mobility (**Paper III** and **Section 4).**

Lastly, an LCA on the C&DW utilisation scenario was carried out with the purpose of providing an overall assessment of this practice, based on findings from the previous phases of the research. Numerous examples of LCA studies exist about residue utilisation (e.g., Coelho and de Brito, 2012; Koletnik et al., 2012; Kucukvar et al., 2014; Loijos et al., 2013; Mercante et al., 2011), though very few of them take into account leaching and toxicity aspects and are thus missing one of the key aspects of LCA, namely comprehensiveness. In addition, detailed contaminant fate in sub-soil has often been disregarded. The results of this part of the work are reported in **Paper IV** and **Section 5** of this synopsis.

2 Materials and methods

2.1 C&DW samples and characterisation

Twenty-nine C&DW samples (C0–C28) were collected in the period 2011-2012 from several Danish recycling facilities handling (i.e. crushing and selling) C&DW. The sampling aimed at being geographically representative of C&DW in Denmark. Despite the focus of this research being primarily on concrete and masonry, often these fractions are found mixed with others, e.g. soil and asphalt, which was reflected in the samples collected for analysis (see Table 3). In addition, four samples of newly cast concrete cores (C29–C32) were obtained in 2012 from pre-cast concrete producers.

All samples were analysed for total content of a wide range of constituents: Al, As, Ba, Ca, Cd, Cl, Co, Cr, Cu, Fe, K, Li, Mg, Mn, Mo, Na, Ni, P, Pb, S, Sb, Se, Si, Sr, V, Zn, total carbon (TC) and total organic carbon (TOC). Additionally, 16 PAH congeners and seven PCB congeners were analysed (details in Butera et al. (**I**).

Table 3 Main characteristics of the 33 C&DW samples analysed during the project.

Sample ID	Origin (Site ID)	Description
C0-C5	A	Clean concrete
C6-C7	B	Construction waste (clean concrete): BASIS[a] + RAPID[b] Cement + fly ash (FA)
C8-C13	C	Mixed aggregates (concrete, bricks, tiles, mortar). Presence of soil, clay
C14-C17	D	Clean concrete
C18-C20	E	Mixed aggregates (concrete, bricks, tiles, mortar). Presence of soil, clay
C21	E	Mixed aggregates (mainly concrete, only few bricks and tiles)
C22-C23	F	Mixed aggregates (mainly concrete, only few bricks and tiles)
C24	F	Clean asphalt
C25-C26	G	Clean concrete
C27-C28	H	Clean concrete. Presence of small metal wires; woods and tiles
C29	I	Newly cast concrete cores: BASIS[a] Cement; no FA
C30-C31	J	Newly cast concrete cores (two different recipes): RAPID[b] Cement; no FA
C32	K	Newly cast concrete cores: RAPID[b] Cement; no FA

[a] BASIS cement is a Portland limestone cement from Aalborg Portland.

[b] RAPID cement is a Portland cement from Aalborg Portland.

2.2 Leaching estimation

The release of constituents into the water phase was investigated by means of leaching tests. The main features of leaching tests for granular materials applied in this work are reported in Box 1, and they are graphically summarised in Figure 1.

Box 1 Description of leaching experiments for materials in granular form, as applied in this project.

Batch test: material in crushed form is mixed with a leachant (typically distilled water) in a bottle at a specific liquid-to-solid (L/S) ratio, and then it is shaken for a relatively short time (24-48h). A simple and fast test, it is often prescribed for compliance purposes, though batch test conditions might not reflect real life in terms of, for example, liquid/solid contact, flow and particle size (López Meza et al., 2010). The standard batch test EN 12457-1 (CEN, 2002) was applied to all 33 collected samples as a first screening of their leaching behaviour, and also for comparative purposes with existing regulations.

pH dependence/static test: material in crushed form is mixed (at a specific L/S ratio) with leachant kept (manually or automatically) at fixed pH values within a range of interest; several bottles, running in parallel, are shaken for a relatively short time (typically 48h). Besides providing information about the acid/base neutralisation capacity (ANC/BNC) of the material, it is applied with the purpose of understanding behaviour at different pH levels, thereby giving indications about geochemical properties, and it serves as a basis for geochemical modelling. The pH static standard procedure EN 14997 (CEN, 2004) was applied to six samples selected based on their batch leaching behaviour.

Percolation test: material in crushed form is compacted into a column and percolated with a leachant (typically distilled water) in up-flow mode under saturated conditions. The purpose is to describe leaching behaviour under percolation conditions while at the same time providing constituent eluate concentrations at increasing L/S values (typically within 0.1 $L \cdot kg^{-1}TS$ and 10 $L \cdot kg^{-1}TS$). The standard column test DIN EN 14405 (CEN, 2014) was applied to six samples.

Lysimeters: large-scale leaching experiments where leachant (in some cases naturally falling rainwater) is gravity-driven through the material and collected at the bottom. They are normally implemented by varying one or more of the conditions recommended by standard column procedures, in an effort to overcome some of the differences between laboratory and field leaching estimations, for example flow conditions, geometry and particle size (typically fixed to standard values in laboratory experiments, to ensure reproducibility and to approach local equilibrium conditions). Lysimeters were applied to six samples with the purpose of assessing the effect of particle size reduction and flow interruptions, and they were continued until approximately L/S 13-14 $L \cdot kg^{-1}TS$.

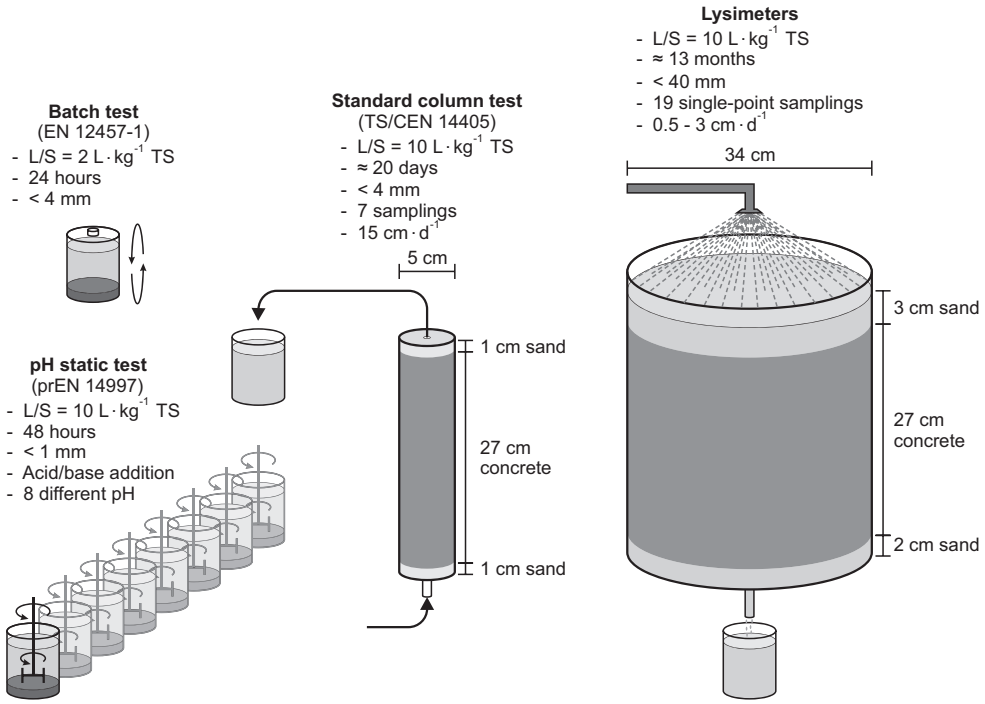


Figure 1 Summary of the leaching tests performed during the study, as reported in Butera et al. (**II**): schematic model of the pH static test, batch test, standard columns and lysimeter, as well as main test features: liquid-to-solid (L/S) ratio, test duration, particle size, Darcy velocity of the leachant and number of samplings (for column and lysimeter tests only).

The mechanisms controlling leaching can be different (namely solubility, availability, diffusion, complexation, sorption to hydrous iron/aluminium oxides (HFO/AlO) and incorporation into other mineral phases (e.g. Chandler et al., 1997; Hykš et al., 2009a; Kosson et al., 1996; Kumarathasan et al., 1990;

Meima and Comans, 1997; Meima et al., 1999; van der Sloot, 1990) depending on the matrix and the constituent, and may be identified through geochemical speciation modelling (see Box 2).

Similarly to MSWI BA, the levels and patterns of elemental release from C&DW may vary upon ageing of the material, owing to carbonation, which is the weathering reaction through which certain minerals may uptake atmospheric carbon – in the case of C&DW, the mineral involved is primarily calcium hydroxide (i.e. portlandite). Upon reacting with carbonic acid (from the dissolution of atmospheric carbon dioxide into concrete pore water), calcium hydroxide is converted to calcium carbonate (i.e. calcite); at the same time, the pH decreases, thus leading to the dissolution of other mineral phases (Bary and Sellier, 2004; Lagerblad, 2006). Carbonation rate and extent are influenced greatly by humidity, temperature and carbon dioxide accessibility, i.e. surface area, porosity and exposure conditions (Engelsen et al., 2005).

Laboratory leaching estimations may be scaled-up to field conditions by means of the liquid-to-solid (L/S) ratio, which is a measure of time in relation to the material's exposure to the leachant (see Hjelmar, 1990). However, a number of challenges exist concerning the relationship between laboratory and field release estimations (see e.g. Kosson et al., 2014), and care should be taken when extrapolating from laboratory to field conditions, especially in relation to redox and pH conditions.

Box 2 Geochemical speciation modelling: conceptual approach.

Geochemical modelling: Based on equilibrium leaching data (typically pH static experiments, though batch data can also be used), measured pH values and measured/assumed redox conditions, as well as on a thermodynamic database of mineral phases, saturation indexes (SI) are calculated for all the mineral phases included in the database. It may be assumed that minerals close to equilibrium with the studied solution control the solution concentration of some elements through dissolution/precipitation. Solution concentrations, calculated based on the SI, can then be compared to the measured ones. After providing information about the available concentration of each constituent, a prediction can be improved iteratively by introducing the effect of other mechanisms, e.g. complexation with organic matter or surface sorption reactions. Literature values for the relevant parameters may be used on first approximation, and then iteratively optimised by fitting model predictions to measured concentrations. Alternatively, selective extractions may be used for estimating humic/fulvic acids and reactive surfaces (HFO/AlO, clay). The

outcome of geochemical modelling is the distribution of the available amount of each constituent between the hypothesised controlling processes: mineral dissolution/precipitation, complexation with dissolved organic carbon (DOC), HFO/AlO sorption. The availability and quality of thermodynamic data affect the modelling results; for example, modelling incorporation of constituents into other mineral phases is not possible if appropriate thermodynamic data for substituted-mineral are not available.
Detailed information about geochemical modelling may be found in e.g. Chandler et al. (1997); Dijkstra et al. (2008); Engelsen et al. (2010, 2009); Meima and Comans (1997); van Zomeren and Comans (2004).

Eluates from the leaching tests were analysed for the same constituents quantified in the solid phase. Quantification of PAHs and PCBs in the eluates, however, was not carried out, owing to their hydrophobic nature. Information on Cr oxidation state in the eluates is valuable in view of the potential toxicity and mobility of emissions. After quantifying the anionic Cr fraction, separated by means of cation-exchange cartridges (Ball and McCleskey, 2003), speciation calculations were performed with the geochemical modelling code PHREEQC (Parkhurst and Appelo, 2013).

2.3 Assessment of behaviour in soil: chromium

Several elements released by C&DW might interact with soil, and their mobility might be affected. During this research the behaviour of Cr in soil was addressed in more detail, owing to the importance of this element in terms of limit values, redox-dependent toxicity and mobility. Batch experiments were carried out by mixing in full factorial design three sub-soils with six types of C&DW leachate obtained from batch testing the collected samples. Total and hexavalent Cr solution concentrations were monitored over approximately six months.

Measured Cr(VI) concentrations in the C&DW leachate were then fitted according to a second-order kinetic model for Cr(VI) reduction. Based on the estimated second-order reduction rate constant and reduction capacity of the sub-soil, Cr(VI) transport in the sub-soil was modelled according to the diffusion/dispersion-advection equation (Trapp and Matthies, 1998).

2.4 Life Cycle Assessment

A life cycle assessment (LCA) is a holistic method for environmentally assessing products or systems. By accounting for environmental exchanges throughout the entire life cycle of the analysed product/system, the LCA approach avoids impact shifting from one life cycle phase to another, or from one environmental compartment to another (Wenzel et al., 1997). As such, the LCA is a valuable tool in the analysis of complex systems, such as waste management systems, and it has been used commonly to assess waste management systems in recent years (e.g. Birgisdóttir et al., 2007; Fruergaard et al., 2010; Laurent et al., 2014a, 2014b; Manfredi and Christensen, 2009). This section only provides a short introduction to LCA, by focusing on aspects that are relevant when residue utilisation is evaluated; details about LCA principles and methodology can be found elsewhere, e.g. EU JRC (2010) and Wenzel et al. (1997).

After defining the goal and the scope of the assessment, as well as the system boundaries, all energy and resource inputs/outputs, as well as waste streams and emissions, are inventoried. It should be noted that emissions are modelled in an LCA in terms of instantaneous cumulative releases into the air, water and soil (hence as amounts as opposed to e.g. concentration) over the considered time horizon (typically 100 years). In the impact assessment phase, characterisation factors (CFs) are used to convert and aggregate the long inventory of environmental exchanges into a relatively few environmental impacts (e.g. global warming, acidification, toxicity to humans or to aquatic ecosystems). Several methods can be used to translate environmental exchanges into impacts (e.g. USEtox (Rosenbaum et al., 2008), ReCiPe (Goedkoop et al., 2013)), and the International Reference Life Cycle Data System (ILCD) recently developed some recommendations concerning the selection of appropriate methods for each impact category (EU JRC, 2011; Hauschild et al., 2013).

The use of an LCA in evaluating scenarios involving leaching from residues (typically released in low concentrations over extremely long timeframes) might, however, be critical (e.g., Allegrini et al., 2014; Hellweg et al., 2005; Pizzol et al., 2011a, 2011b; Schwab et al., 2014) because i) site-specific aspects such as the nature and characteristics of the soil receiving the emissions cannot be captured by such a general method; ii) life cycle impact assessment (LCIA) methods still lack consensus tools for adequately assessing emissions

into compartments such as sub-soil and groundwater, and iii) at the moment offer limited coverage of substances in terms of CFs, especially for inorganic pollutants; iv) ecotoxicological impacts are not a linear function of concentration, as assumed in LCIAs, and therefore should be assessed based on actual concentrations rather than amounts, and v) despite the efforts made by e.g. Hauschild et al. (2008), a well-accepted methodology is still lacking concerning how to handle emissions taking place in the far future and potentially in low concentrations. While other tools such as risk assessment are able to give a site-specific evaluation of impacts by considering local and current conditions, LCA provides a more generic approach to evaluating the overall goodness of the practice at a larger scale, on the one hand, and highlights hotspots in terms of critical processes or substances that might be causes of concern, on the other hand. As such, the two approaches should be considered complementary and not competing (see, for example, Sleeswijk et al., 2003; Tiruta-Barna et al., 2007).

3 Characterisation of C&DW: composition and leaching

One of the aims of this PhD is to provide a database for characterising Danish C&DW. While extensive research has been conducted on the physical, chemical and environmental characterisation of cementitious materials (e.g., Garrabrants et al., 2002; Goodwin et al., 2008; Sanchez et al., 2002; van der Sloot, 2000), the share of literature focusing on real C&DW, rather than on reference materials or laboratory-formulated concretes, is limited and often provides incomplete datasets (e.g., Engelsen et al., 2010, 2009; Galvín et al., 2012; López Meza et al., 2010). In this respect the data produced during this project, reported mainly in Butera et al. (**I** and **II**), provide information concerning the quality and granulometry of C&DW intended for recycling, the content of persistent organic pollutants (POP) of concern (PCBs and PAHs), total content and leaching (according to different testing approaches) of 26 inorganic constituents, carbon (total, organic and dissolved) and pH (thus, indirectly, indication the ageing level of the material intended for utilisation).

3.1 Characterisation of C&DW

Average and 90% confidence intervals for the content and release of analysed constituents are reported in Figure 2 and Figure 3. One of the most evident outcomes of this investigation is the heterogeneity of Danish recyclable C&DW, which is notable especially for minor and trace elements and for the released amounts. The ranges of concentrations found, as reported in Butera et al. (**I**), are in line with previous literature findings for cementitious materials (Bødker, 2006; Delay et al., 2007; Engelsen et al., 2010, 2009; Galvín et al., 2013, 2012; Kalbe et al., 2008; López Meza et al., 2010, 2008; Nielsen et al., 2006; Schiopu et al., 2009; Strufe et al., 2006; van der Sloot, 2000; van der Sloot et al., 2008b; Wahlström et al., 2000; Wehrer and Totsche, 2008).

Composition and contamination

C&DW is mainly constituted by Si (270 $mg \cdot kg^{-1}TS$), Ca (85 $mg \cdot kg^{-1}TS$) and Al (40 $mg \cdot kg^{-1}TS$) – typical for cementitious materials – and has limited organic carbon content (0.4% on average). Chromium, which might derive from raw materials, fuels and additives in cement or concrete production, as well as from contact with grinding equipment during the production phase (Bhatty,

1995), is present in average concentrations of 20 $mg \cdot kg^{-1}TS$, and in all cases it measures below 50 $mg \cdot kg^{-1}TS$.

Despite being banned several decades ago, PCBs can be detected in Danish recyclable C&DW; Butera et al. (**I**) report concentrations of PCB_{TOTAL} in recyclable C&DW in the range 2-70 $\mu g \cdot kg^{-1}TS$. Previous measurements (Jang and Townsend, 2001; Strufe et al., 2006) featured higher limits of detection (LOD) than the current ones, and therefore PCBs were detected only in a few cases, albeit in relatively high concentrations (230-650 $\mu g \cdot kg^{-1}TS$). Our measurements, on the other hand, show for the first time that low but non-negligible levels of PCBs may be found in recyclable C&DW, and even in relatively recent ones, suggesting that these compounds might have spread in matrices different to those where they were actually employed. Additional measurements on cement and concrete raw materials have confirmed background PCB concentrations between 4 and 7 $\mu g \cdot kg^{-1}TS$ in granite, sand and cement (Butera et al. (**I**)), values that essentially confirm previous studies (e.g., Meijer et al., 2003) which found non-negligible background concentrations of PCBs in remote areas. High recycling rates for C&DW, generally perceived as a success by authorities, might produce adverse consequences in environmental compartments not previously affected by these compounds, if PCB-contaminated C&DW enters the recycling scheme. Even though the PCB levels found during this study were not critical (as discussed in section 3.3), the presence of POPs in recyclables is a topic of great concern, owing on the one hand to their potential accumulation in secondary materials (e.g., Pivnenko et al., 2014 for recycled paper), and on the other hand to uncontrolled spread into different environmental compartments.

Leachability

Eluates from C&DW batch leaching tests show pH values between 10 and 13, thus exhibiting a wide range of ageing conditions, although complete carbonation was not reached by any sample. The presence of masonry in the samples, however, may contribute to lowering the pH value, owing to the less alkaline nature of clay-based materials.

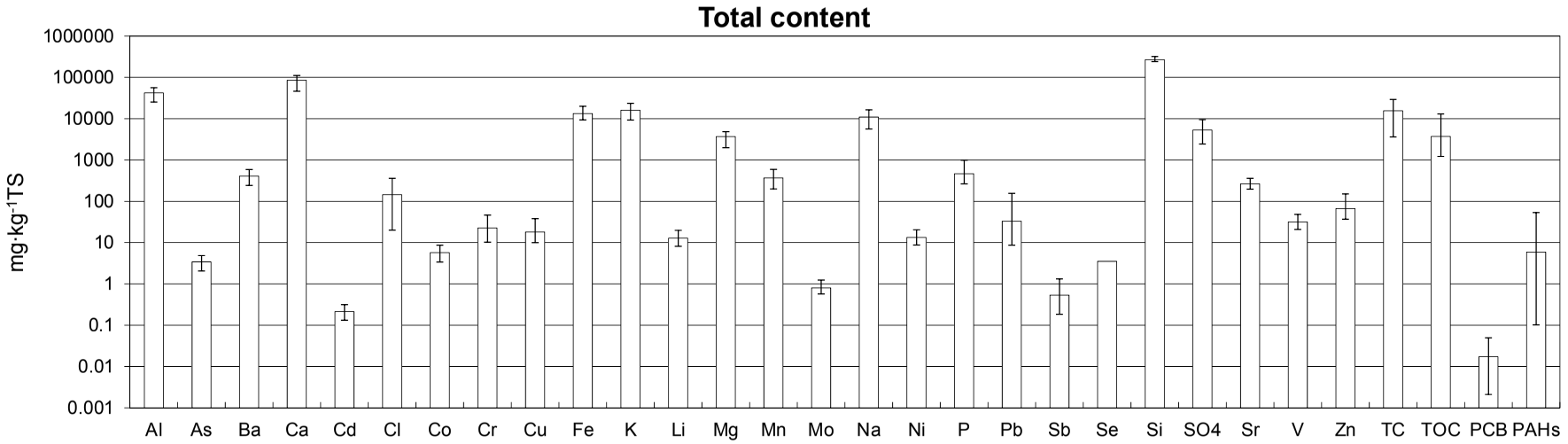


Figure 2 Variability of the measured total concentrations for the 33 samples (average and 90% confidence intervals). Selenium was in all cases below limit of detection (LOD) of 7 $mg \cdot kg^{-1}TS$. PCBs: PCB_{TOTAL}, calculated as PCB_7x5. PAHs: sum of 16 EPA-PAHs. Data are based on Butera et al. (**I**).

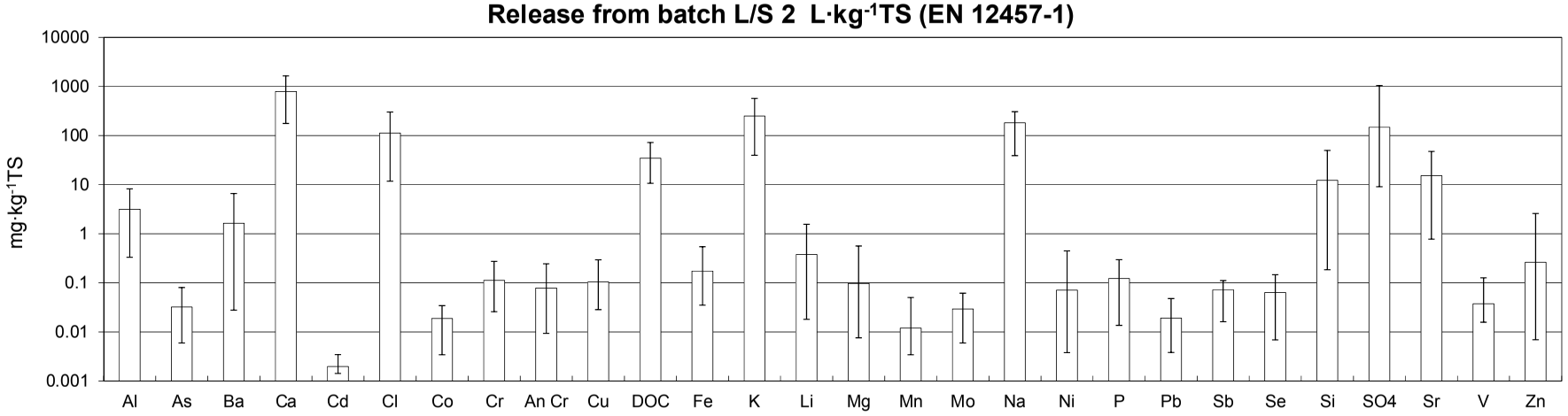


Figure 3 Variability of the measured released amounts for the 33 samples (average and 90% confidence interval). Data are based on Butera et al. (**I**). Cd was in most cases below LOD, while As, Fe, Pb, Sb and Zn, in several cases, were below LOD or the limit of quantification.

As shown in Figure 3, eluates from C&DW are dominated by Ca and sulphate, as well as anions from the dissolution of soluble salts (e.g. K, Na and Cl^-). In terms of trace elements, chromium was found in the range 5-360 $\mu g \cdot kg^{-1}$TS, closely followed by the values for anionic chromium. Approximately 80% of Cr was found in anionic form, which may be interpreted as hexavalent species, mainly chromate, based on speciation calculation (Butera et al. (**I**)). This appears reasonable considering the process of concrete production (mildly oxidising conditions in the cement kiln) and the negligible presence of DOC, a potential reductant for Cr(VI).

Interestingly, large inter-sample differences (Ca, Ba, Cl^-, Cr, K, Li, Mg, Na, Sr, Se, Si, SO_4 and V) could be correlated with observed differences among the samples in terms of pH (see Figure 4). The latter, in turn, may be interpreted as a result of moderate ageing, but it may also be related to the presence of masonry in some of the studied samples (as highlighted in Table 3). As described in Butera et al. (**I**), samples with lower pH exhibited higher SO_4, Si, Cr, Mg, Cl^-, V and Se leaching and lower release of Ca, Sr, Ba, Na, K and Li. Such behaviour is typical of carbonated residues, and it reflects the mineralogical changes taking place upon carbonation, namely portlandite and ettringite dissolution leading to the release of sulphate-substituting ions such as chromate and vanadate; the progressive decalcification of calcium-silicate-hydrate (CSH) phases into more soluble amorphous silica phases; the precipitation of calcite and the dissolution, for example, of barium and strontium sulphates with the formation of less soluble carbonates (Garrabrants et al., 2004; Müllauer et al., 2012; Van Gerven et al., 2003). However, data in the literature (Alonso-Santurde et al., 2010; Galvín et al., 2013; Strufe et al., 2006) reveal that bricks might present similar leaching patterns. Although source segregation is a prerequisite in Denmark, clean fractions might end up being mixed, since the utilisation of a blended product has technical advantages over the use of clean bricks alone (Hansen and Lauritzen, 2004). Based on the results of this study, however, the production of a clean concrete fraction is preferable from an environmental standpoint. If crushed bricks are not suitable for utilisation, their reuse should be promoted: aside from practical and logistical difficulties, a recent life cycle assessment study found brick reuse to be more environmentally friendly than utilising them as a filling material in crushed form (Møller et al., 2013).

Increasing the carbonation rate before final utilisation, by means of enhanced aeration, is an option that is sometimes proposed with the purpose of maximising CO_2 uptake from C&DW and therefore improving the carbon foot-

print of cementitious materials. However, the effect of such pre-treatments on leaching behaviour should be taken into account in a comprehensive manner, considering that even a moderate carbonation degree might be associated with substantial changes in leached amounts (see Section 5).

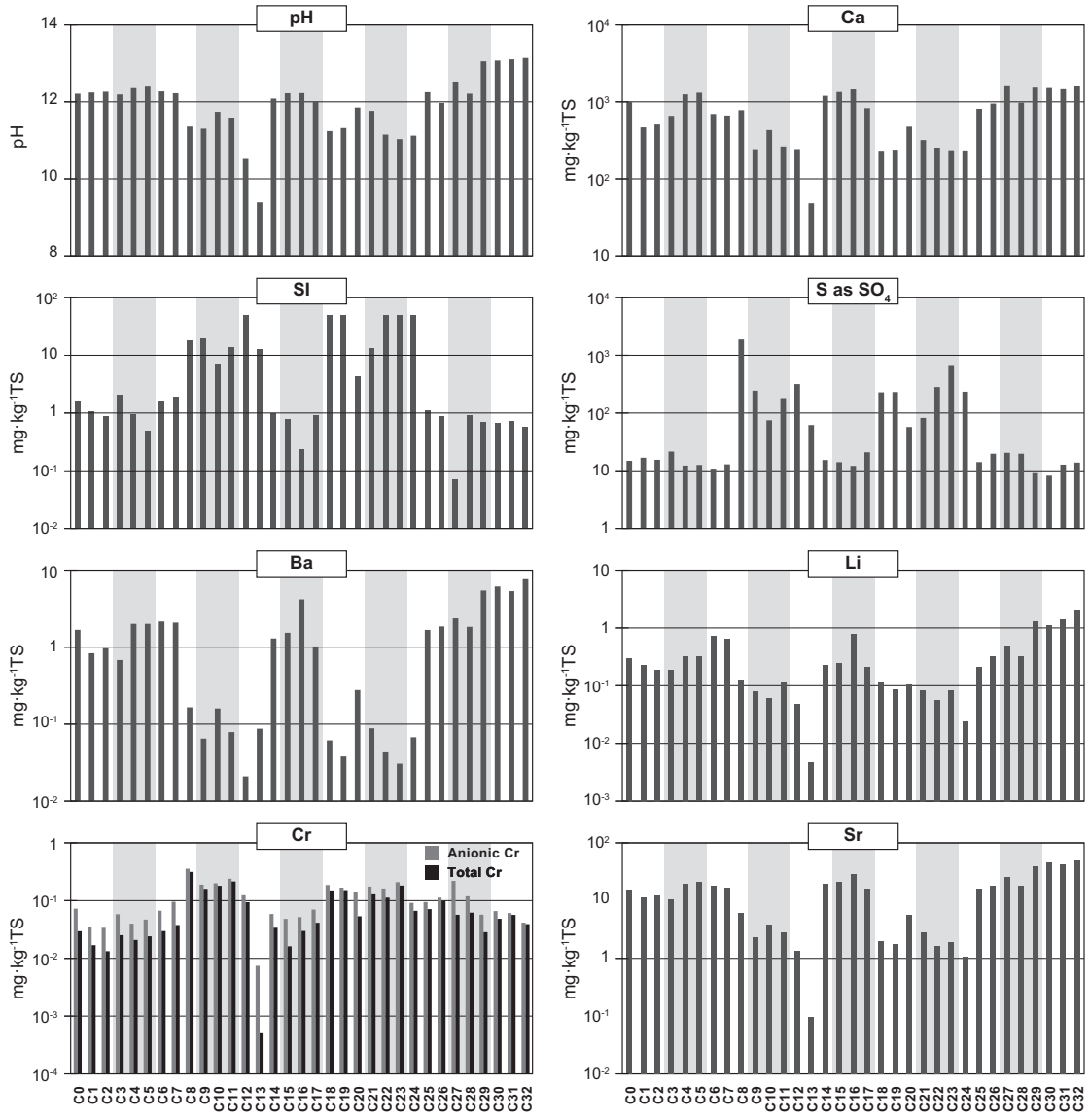


Figure 4 Leaching behaviour of the 33 samples for pH and selected elements after the batch EN 12457-1 L/S 2 test (CEN, 2002), from Butera et al. (**I**). Each bar represents the average of at least two replicate measurements. Significant differences can be noted between samples C8-C12, C18-C23 and the remaining samples.

3.2 Percolation experiments

Field testing should always be preferred to laboratory experiments, due to the different conditions that may occur, especially in terms of redox (Kosson et al., 2014); however, field investigations are often impractical, lengthy and costly, so assessment generally relies on laboratory experiments, with the most relevant test type for percolation conditions being the up-flow column percolation test (e.g. DIN EN 14405, see Box 2). The representativeness of laboratory leaching tests, however, is often questioned in relation, among others, to two critical parameters, namely particle size and flow conditions. C&DW is normally utilised in grain sizes up to 40mm, though the conditions imposed in standard column tests in order to approach local equilibrium, and hence reproducibility (saturated upward flow and a maximum particle size of 4mm), might lead to major issues. Nonetheless, data reported in Butera et al. (**II**) demonstrate that when the focus is on quantifying the *cumulative release at L/S 10 L·kg^{-1}* from C&DW, this is not the case for most analytes of interest. The comparison between cumulative release at L/S ratio of 10 L·kg^{-1} from standard up-flow percolation and down-flow lysimeters with coarse-grained material, summarised in Figure 5, shows that release estimates are within a factor of 2 in 80% of the cases. The release of P, and to a lesser extent Ba, Mg and Zn, is consistently lower in standard columns, and the release of Pb is higher. However, more differences are observed in terms of concentrations observed in the early leachates, in that Al, As, Ba, Cd, Cu, DOC, Mg, Mn, Ni, P, Pb, Sb, Se, Si and Zn exhibit differences up to over a factor of 100.

Initial pH values were higher in standard columns, likely owing to grain size reduction resulting in the exposure of fresher surfaces, which might in turn have affected the leaching behaviour of those elements showing differences.

Results from Butera et al. (**II**) partly confirm previous research (Delay et al., 2007; Hjelmar et al., 2013), though any comparison with literature findings is not straightforward, since this is one of the first studies focusing on the effect of flow conditions and particle size on C&DW release estimates for a large set of analytes. Most of the existing studies have focused on one aspect at a time, thus making the outcomes not fully comparable. For example, López Meza et al. (2009) concluded that different types of intermittent flow do not significantly affect C&DW release estimations compared to saturated standard columns. Furthermore López Meza et al. (2010) also studied the effect of different column contact times on the release of copper, chromium, sulphate

and chloride from C&DW, concluding that variations in contact time did not have a strong effect on their leaching behaviour. However, the authors focused on the effect of even shorter contact times than required in standard tests (in order to improve further the practical feasibility of the procedure), rather than on the effect of laboratory-accelerated tests compared to slower and more realistic flow conditions. On the contrary, Wehrer and Totsche (2008), who focused on the effect of flow velocity and interruptions on As, Cl^-, Cd, Cr, Cu, DOC, Ni, Pb and Sn release, concluded that at the flow rate recommended by standard up-flow percolation procedures, non-equilibrium conditions may exist and affect the release of DOC, Cl^-, Cr, Cu and Ni. However, neither of these studies included the effect of particle size in their assessment. Hjelmar et al. (2013) showed that different particle size affected the release of As, Fe and Zn from masonry materials, while Ba and Mg showed good agreement, though their values for Al, Cu, DOC, Mg, Mn, P, Pb, Sb, Se, Si (i.e. the other elements showing differences between the two percolation setups of Butera et al. (**II**)) were too low to draw any conclusions. Galvín et al. (2014) concluded that a higher share of fine particles might increase Cr and SO_4 leaching from C&DW, while Delay et al. (2007) compared standard column tests and intermittent down-flow lysimeters for a C&DW reference material (particle size of 4mm in both cases), finding good agreement for most analytes (though only Ca, Cl^-, Cr, Cu, K, Na and SO_4 were analysed, and only until L/S 0.6 $L \cdot kg^{-1}TS$).

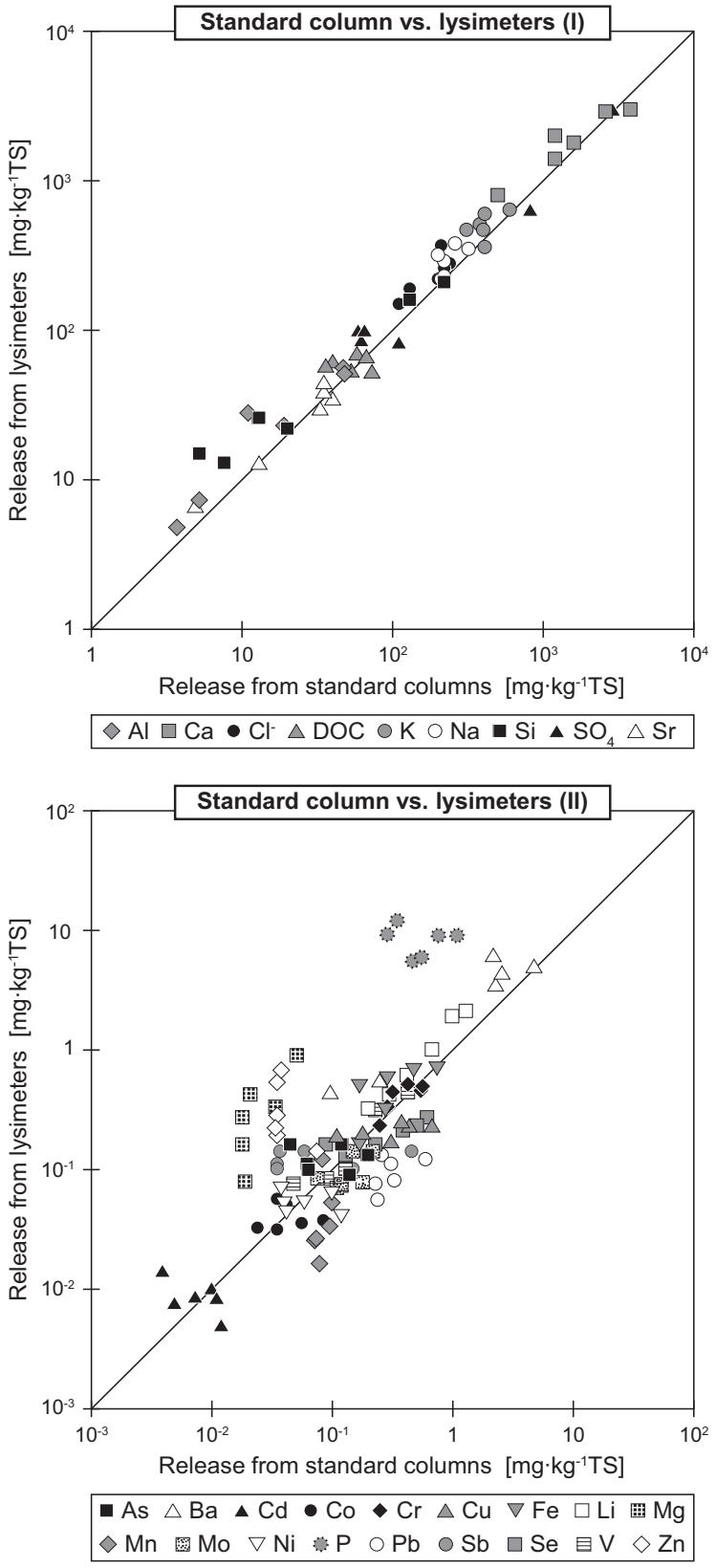


Figure 5 Comparison between standard column tests and lysimeters for all analysed elements (Butera et al. (**II**)).

3.3 Critical parameters

The release and concentration values obtained through batch and column testing may be put into perspective by comparing them to existing limit values. It should be noted, nevertheless, that most of the regulations used are not strictly applicable to C&DW, and therefore they are used here only for comparison; furthermore, no risk assessment was carried out during this study. The following comparison represents a basic identification of the potentially critical elements. Regulations such as waste acceptance criteria (WAC) (EU Landfill Directive 2003/33/EC, European Commission, 2002) are developed with the purpose of protecting groundwater resources (Hjelmar, 2012), and therefore they might provide an indication of the level of concern associated with emissions.

Based on the comparison with different national limit values for utilisation of secondary materials in construction applications, as well as the European WAC, reported in Butera et al. (**I**, **II**), the most critical inorganic parameters appear to be Se, Cr, Sb, SO_4 and Cl^-. These findings are in agreement with previous studies on C&DW (van der Sloot, 2000; Wahlström et al., 2000). In particular, while selenium has not been object of extensive research so far, chromium has been regarded for a long time as an element of concern in cementitious materials, in particular in its hexavalent form. As an example, reducing agents have been added in some cases to cement or concrete, in order to protect workers' health from soluble Cr(VI). On the other hand, typical elements of concern in MSWI BA (such as Cu, Pb, Zn) are released by C&DW in relatively lower amounts, and often below detectable levels (especially Pb and Zn).

Based on the results from Butera et al. (**I**), carbonated and masonry-containing samples appear more critical than fresher and unmixed C&DW (i.e. clean concrete), because of higher leaching of regulated substances, e.g. sulphate, chromium, chloride and selenium. This aspect should be taken into account when evaluating practices for C&DW management.

Organics are assessed based on their total content rather than on their leachability. PCBs are well below the requirement of the Dutch Soil Quality Decree (SQD) (Dutch Ministry of Housing Spatial Planning and the Environment, 2007) and the limit for inert landfills (EU Landfill Directive). Similarly, PAHs in C&DW are below SQD limit values, except for one asphalt sample.

4 Fate of Cr(VI) and other pollutants

The fate of inorganic pollutants in the subgrade, below the road application of C&DW, is an important aspect to consider when assessing the overall goodness of the reuse practice, and therefore it has been addressed during this PhD project.

Sorption is a key mechanism at the heart of interactions between inorganic pollutants and soil (see Box 3). Over the years, soil sorption has been studied extensively from both a theoretical (e.g. Bradl, 2004) and an applied point of view (e.g., risk assessment (Dijkstra et al., 2009, 2008), LCAs (Allegrini et al., 2014; Schwab et al., 2014), establishment of regulatory frameworks (Verschoor et al., 2008)).

Box 3 Sorption of metals to soil: key concepts (Bradl, 2004).

The term "**sorption**" indicates the loss of a metal ion from an aqueous to an adjacent solid phase and consists of three important processes: adsorption, surface precipitation and absorption.

Adsorption is a two-dimensional accumulation of matter at the solid/water interface related to intermolecular interactions between solute and solid phases. Non-specific adsorption or ion exchange is an electrostatic phenomenon involving rather weak and non-selective outer-sphere complexes whereby cations from the pore water are exchanged for cations near the surface. Specific adsorption is characterised by more selective and less reversible reactions (inner-sphere complexes): metal ions bind strongly to functional groups at the soil surfaces, typically negatively charged at high pH (hydroxyl (-OH), typically on Fe and Al oxides; clay minerals; carboxyl (-COOH), carbonyl and phenolic groups, typically on soil organic matter; colloidal particles).

Surface precipitation involves the three-dimensional growth of a new solid phase. Metals may precipitate onto soils as oxides, hydroxides, carbonates, sulphides and phosphates. Surface precipitation is mainly a function of pH and the relative quantities of metals and anions present.

Absorption involves the diffusion of an aqueous metal species into the solid phase, and therefore it is three-dimensional in nature. Heavy metals that are specifically adsorbed onto clay minerals and metal oxides may diffuse into the lattice structures of these minerals, thus becoming fixed into the pore spaces of the mineral structure (solid-state diffusion).

Although several substances are subject to interactions with soil particles, the behaviour of Cr in soil appeared of specific importance and was assessed with a greater level of detail. Chromium was among those substances closer to existing limit values; furthermore, its interactions with soils are made more complex by its redox behaviour. Hexavalent Cr, which is toxic, mutagenic and carcinogenic, significantly soluble and thus mobile (Barceloux, 1999; von Burg and Liu, 1993), might, upon interaction with soil particles, be reduced to trivalent chromium, which is a nutrient (Anderson, 1997), and sorbs strongly to soils (Weng et al., 1994) and has low solubility above pH 5.5 (Bartlett and Kimble, 1975) – and therefore limited mobility. As a result, the fate of Cr(VI), after its release from C&DW utilisation in road construction, is an essential aspect to consider when evaluating the actual hazardousness of emissions, and it was addressed experimentally. For other inorganic elements, soil interactions were addressed based on the literature, for the purpose of the LCA study (see Section 5.1).

4.1 Cr(VI) reduction in soil

Based on the experiments described in Butera et al. (**III**), and summarised in Section 2.3, some degree of retention of Cr(VI) in alkaline C&DW leachate in contact with organic matter-poor soils is observed: 30% to 99.6% of initial Cr(VI) was removed from the leachate after six months of contact time. Different combinations of soils, initial Cr(VI) levels (selected based on lysimeter experiments reported in Butera et al. (**II**)) and leachate types might lead to different retention kinetics (Figure 6); however, in all cases, the time-dependent evolution appeared too slow to be explained by adsorption processes, which typically take place over a few hours to a few days (e.g., Zachara et al., 1989). It is hypothesised that a reduction to trivalent form might be responsible for the observed immobilisation, with soil organic matter (SOM) as a possible reductant, although an effect from Fe(II) cannot be excluded. The experiments also revealed that some Cr(III) might be found in solution, most likely through complexation with SOM.

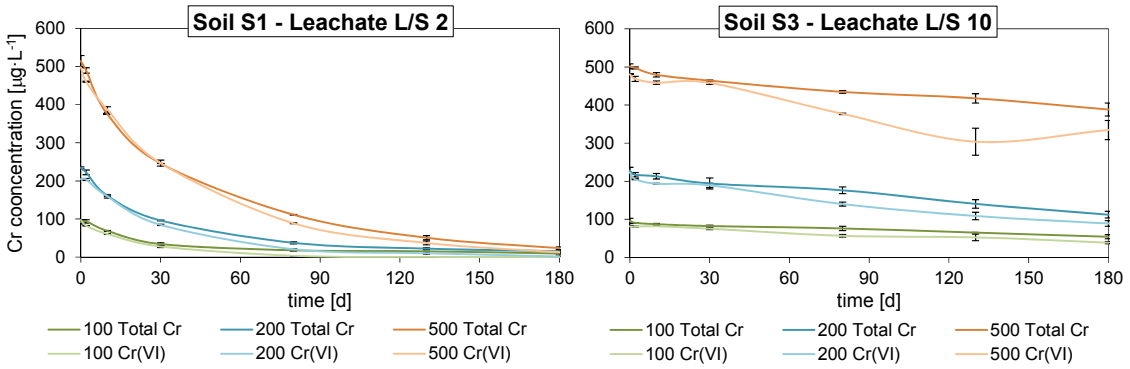


Figure 6 Two examples of total and hexavalent chromium concentration profiles over time for different combinations of soil and leachate (2 L/S ratios: L/S 2, L/S 10 $L \cdot kg^{-1}$; 3 initial Cr(VI) concentrations: 100, 200, 500 $\mu g \cdot L^{-1}$). Each data point corresponds to the average of two replicate samples; individual measurements are shown as the maximum and minimum of the error bars. Adapted from Butera et al. (**III**).

4.2 Cr(VI) migration modelling

Knowledge about reduction kinetics and capacity needs to be coupled with information concerning the vertical transport of water, in order to assess the combined effect of the two phenomena on the overall Cr(VI) mobility.

A second-order model could be used to satisfactorily fit the experimental curves of Cr(VI) reduction by sub-soils (see an example in Figure 7). The reaction velocity is proportional to Cr(VI) concentration ($\mu eq \cdot L^{-1}$) and initial reduction capacity of the soil R_0 ($\mu eq \cdot L^{-1}$) through a second-order reaction rate k ($L \cdot \mu eq^{-1} \cdot d^{-1}$). The reduction capacity of the soil represents the theoretical potential of the soil to act as an electron donor for reducing oxidised compounds – the general term "reduction capacity of the soil" is here used to describe the effect of different (and not necessarily identified) electron donors present in the soil.

Cr(VI) migration could then be modelled according to the diffusion/dispersion-advection equation (the fundamental equation describing transport in a porous media), with a loss modelled according to the second-order reduction reaction previously identified (Trapp and Matthies, 1998). The equation was then solved numerically for a time horizon of 100 years. This model includes a description of the source term based on the results from the five lysimeters described in Butera et al. (**II**).

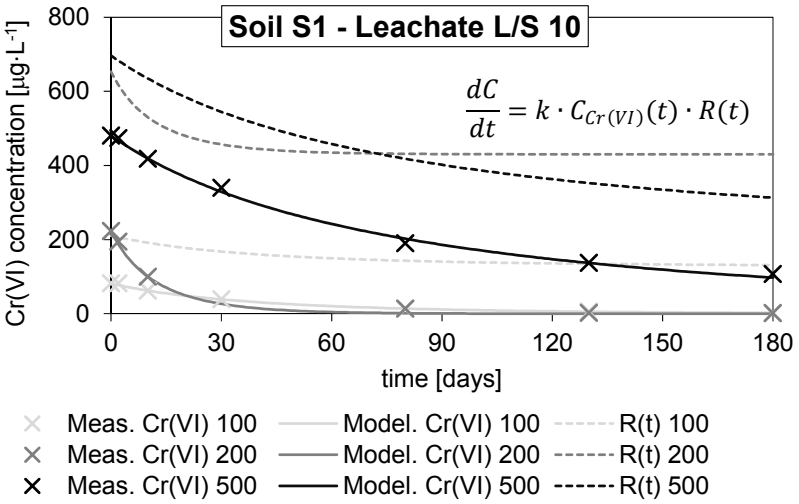


Figure 7 Example of fitting experimental reduction data with second-order model (equation reported) for the case of soil S1 in contact with leachate (produced at L/S 10 $L \cdot kg^{-1}$ TS) at different initial Cr(VI) concentrations. Adapted from Butera et al. (**III**).

In the worst case scenario, which includes the highest release from the five C&DW lysimeters and the slowest kinetics from the 18 sub-soil leachate batch experiments, the results reveal significant Cr(VI) concentrations[1] only in the first 70 cm of sub-soil, and only for the first years after building the road (see Figure 8a). After approximately 40 years, i.e. roughly the lifetime of a road[2] (Birgisdóttir, 2005; Stripple, 2001), dissolved Cr(VI) concentrations were below 20 $\mu g \cdot L^{-1}$ at all depths, decreasing to a maximum of 10 $\mu g \cdot L^{-1}$ after 100 years. Calculating the cumulative release of Cr(VI) at a 1 m depth below the C&DW layer resulted in retention of over 99.99% Cr(VI) by the soil in the worst case scenario, comparable with transfer coefficients calculated by Allegrini et al., 2014; Schwab et al., 2014.

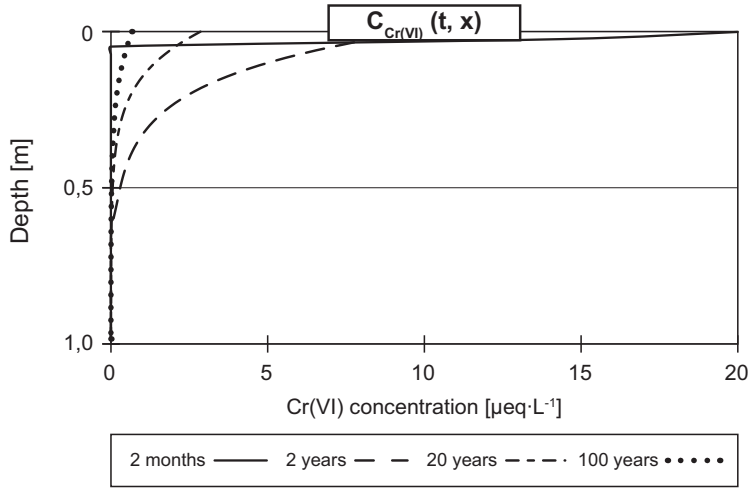
As shown in Figure 8b, the reduction capacity did not deplete during the 100-year time horizon, as slow reduction kinetics were the main limiting factor preventing a complete Cr(VI) reduction. In order to contain Cr(VI) migration further, it is important to minimise rainwater infiltration during both the road

[1] The term "significant Cr(VI) concentrations" is here used to indicate levels above 1 $\mu g \cdot L^{-1}$, a value comparable to the Danish guideline value for Cr(VI) in groundwater: 1 $\mu g \cdot L^{-1}$ (Danish Ministry of Environment, 2014)

[2] While the asphalt wearing course is subject to more frequent maintenance, and is normally replaced every five years, the lower layers (binder, base and sub-base) are designed with a lifetime of approximately 40 years in mind, after which the overall stability of the road needs to be assessed.

construction phase and the utilisation phase, since this will increase vertical water movement and outbalance the reduction effect.

a)



b)

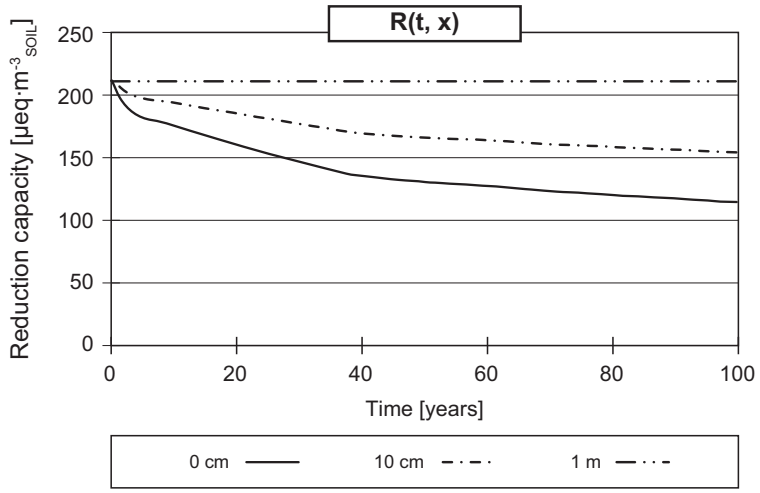


Figure 8 a) Cr(VI) concentrations in sub-soil pore water as a function of depth, at different times and for the worst case scenario. b) Reduction capacity of the sub-soil as a function of time, at different depths and for the worst case scenario. Adapted from Butera et al. (**III**).

Sensitivity to model parameters

Effect of Temperature: Cr(VI) reduction experiments reported in Butera et al. (**III**) were conducted at 25^{0}C, though it should be noted that lower subsurface temperatures may be expected in temperate climates. This aspect may affect Cr(VI) reduction rates, which are sensitive to temperature changes (Elovitz and Fish, 1994; Wittbrodt and Palmer, 1996). Reaction rates at 13^{0}C, estimated through the Arrhenius equation, result in a dramatic increase in Cr(VI) migration, with significant concentrations (> 1 µg·L^{-1}) expected in the

first 180 cm of sub-soil in the first years after release (worst case scenario). However, the transfer coefficient to soil at a 1m depth only decreases from 99.99% to 99.7%.

Infiltration rate: Similarly to temperature, an increase in the infiltration of precipitation (from 10% to 30%), leading to faster pore water velocity, resulted in a considerable increase in Cr(VI) migration, and in the worst case scenario, the first 190 cm of sub-soil was found to be affected by significant concentrations (> 1 $\mu g \cdot L^{-1}$) in the first years after release.

C&DW layer thickness: Cr(VI) migration results were not sensitive to an increases in C&DW layer thickness from 30 to 70 cm.

Environmental significance

Based on the above discussion on model parameters, it appears that in some cases C&DW utilised in road construction might result in significant Cr(VI) concentrations (>1 $\mu g \cdot L^{-1}$) reaching the groundwater table, especially in temperate climates and/or in the presence of elevated infiltration rates. Some key aspects can be identified, which should be addressed on a case-by-case basis, in order to minimise the impacts of this type of application. First, the minimisation of *rainwater infiltration* in road sub-bases is an important aspect: on the one hand, frequent maintenance of the asphalt layer during the road's life is required, and on the other hand water infiltration should also be minimised during the road construction phase. Additionally, the use of C&DW sub-bases in unpaved roads, or under draining asphalt wearing courses, might be critical and should be assessed carefully. As opposed to the thickness of the C&DW layer, other road design parameters, such as *distance to groundwater*, might be critical. The depth of shallow groundwater varies greatly, even within the same region, and therefore a site-specific assessment is recommended. Utilisation of C&DW in an area with a relatively shallow phreatic zone might not be considered appropriate. Finally, modelling based on the C&DW sample showing the lowest Cr(VI) releases among the five studied in Butera et al. (**II**) resulted in migration below 1 m, even in cold climates, thus demonstrating that the *quality of C&DW* might have an effect, at least to some extent. However, compliance with the leaching limit values does not necessarily guarantee the absence of environmental issues; for instance, the worst case scenario analysed in Butera et al. (**III**) included C&DW lysimeter leaching data that were below all existing limit values in terms of cumulative release. In such a scenario, and in temperate climates, however, Cr(VI) might

migrate as deep as 180 cm under the road sub-base, which in some cases might imply the contamination of shallow groundwater.

However, even in a case of significant Cr(VI) migration, the actual environmental consequences should be evaluated objectively: the immission of Cr(VI) into groundwater in concentrations above 1$\mu g \cdot L^{-1}$ might not necessarily be critical, particularly when considering that the Danish guideline value for *Cr(VI) in groundwater* corresponds to 1 $\mu g \cdot L^{-1}$ (Danish Ministry of Environment, 2014). Leachate is subject to significant dilution when mixing with a greater groundwater flow. Furthermore, reducing conditions are expected in the aquifer, and Cr(VI) might not be a problem in the long run. Finally, it should be kept in mind that shallow groundwater is normally not used as a drinking water resource.

One last aspect that is worth discussing is the possibility of Cr(III) re-oxidation (e.g., Bartlett and James, 1979; Fendorf, 1995). Owing to the non-anaerobic experimental conditions, the observed retention of Cr(VI) should already be considered as a net value between Cr(VI) reduction and Cr(III) re-oxidation. Another aspect, however, is Cr(III) complexation with SOM, resulting in increased mobility and Cr(III) potentially reaching environmental compartments (e.g. natural waters) where different conditions might favour its oxidation. While reduced conditions, prevalent in groundwater, would limit such a risk (Lindberg and Runnells, 1984; Takeno, 2005), Cr(III) in oxygenated environments such as surface water bodies could potentially be oxidised by dissolved oxygen or Mn oxides (Fendorf, 1995). Nevertheless, Schroeder and Lee (1975) reported that Cr(III) could only be oxidised at slow rates by O_2 in natural waters, while oxidation by MnO_2 was inhibited in natural waters (owing, for example, to the sorption of Cr(III) on suspended solids, or the sorption of other cations on MnO_2 surfaces). Additionally measured dissolved Cr(III) concentrations, already relatively low (16 $\mu g \cdot L^{-1}$ on average), are expected to be somewhat diluted along the pathway. Overall, it can be concluded that the environmental risk associated with Cr(III) re-oxidation is quite limited.

5 Life Cycle Assessment

The management system for C&DW is a complex system characterised by different and non-comparable environmental exchanges (e.g. savings in virgin aggregates for road construction, emissions related to transportation and to leaching) happening in different places. Life cycle assessment can be particularly useful in analysing such a system in a systematic way, similarly to what has been done for other mineral residues (typically MSWI BA) by, for example, Birgisdóttir et al. (2007). Butera et al. (**IV**) performed an LCA of two end-of-life options for C&DW (namely utilisation in road sub-base structures and landfilling), including leaching in a detailed way, for the first time addressing in detail the fate of emitted compounds in sub-soil.

5.1 Modelled scenarios

The two scenarios modelled within the EASETECH waste LCA framework (Clavreul et al., 2014) had as a functional unit the management of 1 Mg of C&DW:

a) Utilisation of 1 Mg C&DW in road construction, as a filler material in road sub-bases;
b) Disposal of 1 Mg C&DW in an inert landfill.

The scenarios comprised all phases after C&DW generation until the final disposal of all residues, including several transportation steps, leaching from C&DW, avoided emissions from the displacement of primary aggregates (extraction, transportation and leaching) as well as capital goods and operational energy requirements for both the C&DW crushing plant and the landfill. Figure 9 provides a sketch of both scenarios, while more details can be found in Butera et al. (**IV**)

Modelling was carried out according to a consequential approach (see EU JRC, 2010, for details), for a 100-year time horizon and for Danish conditions in relation to climate, marginal technologies and type of sub-soil. The assessed categories were selected based on recommendations from ILCD (Hauschild et al., 2013) namely global warming (GWP), acidification (AP), eutrophication in freshwater, marine and terrestrial environments (FEP, MEP, TEP), ozone depletion in the stratosphere (ODP), ionising radiation (IR), particulate matter formation (PM), photochemical ozone formation (POF), resource depletion of fossil fuels and elements (ADP_F, ADP_E), human toxicity, carcinogenic and non-carcinogenic (HT_C, HT_{NC}) and toxicity to freshwater ecosystems (ET_{FW}).

a)

b)

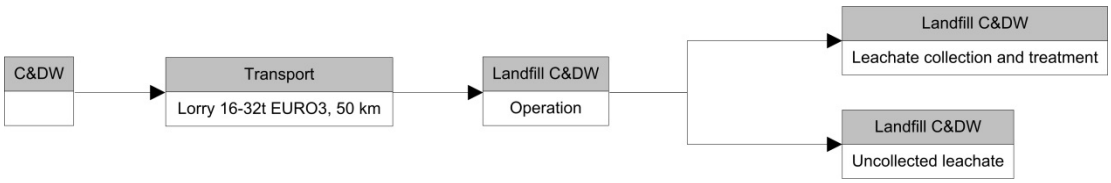


Figure 9 Schematic representation of the road construction (a) and landfill (b) scenarios for C&DW end-of-life management, as modelled in EASETECH. From Butera et al. (**IV**).

In both scenarios, leaching data for C&DW were modelled based on the lysimeter experiments described in Butera et al. (**II**); however, there were major differences between scenarios a) and b) in terms of the L/S ratio reached at 100 years, owing to the different structures of the two configurations (see table 4). Furthermore, landfill leachate is assumed to be collected and treated.

Table 1 Summary of the characteristics of the road structure and of the C&DW landfill, based on Butera et al. (**IV**).

		C&DW/gravel in road sub-base	C&DW in landfill
Thickness	[m]	0.3	10
Density	[kg·m^{-3}]	1850	1500
Net infiltration	[mm·year^{-1}]	0.078	0.4 for 2 years (operation) 0.2 for 98 years (aftercare)
L/S	[L·kg^{-3}]	14.1	1.36

Modelling of the leached pollutants was done taking into account sorption to the sub-soil, as discussed in Section 4. For Cr, the calculated transfer coefficient to soil in the worst case scenario, including the effect of temperature, was used (99.7%, see Section 4), and for the other inorganic compounds fate modelling in soil was accounted for by assuming transfer coefficients based on previous work on similar materials (Allegrini et al., 2014, see Table 5). Table 5 shows that most of the emitted inorganic compounds would be sorbed to the sub-soil and would not represent an issue from a 100-year perspective. The small fraction which is dissolved in pore water at 1m below the road sub-base layer (assumed as the level of the shallow groundwater) is then modelled as being emitted into the freshwater compartment (owing on the one hand to the absence of CFs to the groundwater, and on the other hand to the fact that

shallow groundwater cannot be considered a "closed" compartment but will eventually end up in a surface water body). However, the fraction sorbed to soil cannot be assigned CFs for emissions into soil in USEtox, and so CFs for emissions into water have to be used instead (Schwab et al., 2014), because fate modelling in soil has already been accounted for by the transfer coefficients, and applying CFs for emissions into soil would result in i) double counting of the fate in soil and ii) the inclusion of pathways such as soil erosion, surface runoff, and for human toxicity ingestion or dermal contact, which might be relevant for topsoils but not for sub-soil. ET_{FW} does not include effect factors for species in soils/sub-soils (being a *freshwater*-ecotoxicity category), and the only pathway from sub-soils to humans is through shallow groundwater (i.e. freshwater). Therefore, the fraction remaining in soil also has to be modelled as an emission into freshwater, though its impacts should be regarded as "stored" freshwater and human toxicity.

Table 5 Portion of released metal remaining in the first 1m-deep sub-soil layer below road application of MSWI BA (Allegrini et al., 2014), based on reactive transport calculations in three types of soil (sand, peat and clay) and modelled in LeachXS™-Orchestra (van der Sloot et al., 2008a), using data from five percolation leaching tests on MSWI BA as an input. Based on the similarity of characteristics and application, values for MSWI BA are considered applicable to C&DW utilisation scenarios. The intervals represent the minimum and maximum among the three soil types.

As	100%	-	100%	**Pb**	98.5%	-	100%
Ba	96.4%	-	100%	**Sb**	36.8%	-	57.3%
Cd	94.7%	-	99.8%	**Se**	99.9%	-	100%
Co	97.5%	-	100%	**Sn**	97.4%	-	100%
Cu	100%	-	100%	**V**	93.5%	-	100%
Mo	100%	-	100%	**Zn**	100%	-	100%
Ni	96.7%	-	100%				

5.2 LCA results

LCA results (in Table 6) show that utilisation of C&DW in road construction as a replacement for virgin aggregates is preferable to C&DW landfilling for most impact categories. Differences between potential impacts in the two scenarios are significant (one order of magnitude), apart from HT_C, which shows comparable impacts. Although utilisation of C&DW does not result in environmental savings, in most categories scenario (a) performs better than landfilling (b) thanks to the substitution of virgin aggregates, which does not take place in the landfill scenario. Nonetheless, for three categories (FEP, HT_{NC} and ET_{FW}) the landfilling of C&DW results in lower potential impacts,

owing to lower leaching (lower L/S ratio in the landfill, leachate collection and treatment).

Table 6 Characterised impacts for the analysed impact categories, for both scenarios. Values in % represent percentage variations in total impacts by using 5th and 95th percentile leaching data compared to the scenario with average leaching data, for the categories affected by leaching data, i.e. FEP, HT_C, HT_{NC} and ET_{FW}. Values in bold indicate the scenario with the greatest environmental impacts. From Butera et al. (**IV**).

		Scenario a): C&DW in road			Scenario b): C&DW to landfill		
GWP	$kgCO_2$-eq		8.73E+00			**1.79E+01**	
POF	kgNMVOC-eq		1.32E-02			**1.19E-01**	
PM	kgPM2.5		1.21E-03			**5.94E-03**	
AP	AE		2.37E-02			**1.02E-01**	
FEP	kgP-eq	**-31%**	**8.84E-03**	**30%**	-22%	4.42E-04	47%
MEP	kgN-eq		4.31E-03			**4.00E-02**	
TEP	AE		4.90E-02			**4.41E-01**	
ADP_F	MJ		1.45E+02			**2.58E+03**	
ADP_E	kgSb-eq		7.91E-05			**3.64E-04**	
HT_C	CTU_h	-45%	1.33E-07	**63%**	**-2%**	**1.61E-07**	2%
HT_{NC}	CTU_h	**-40%**	**5.88E-06**	**37%**	-2%	1.12E-06	4%
ET_{FW}	CTU_e	**-49%**	**9.90E+01**	**73%**	-24%	1.09E+01	40%

Important processes and substances

Figure 10a shows that in scenario a) the impact categories can be divided into some which are dominated by transport-related processes (GWP, ODP, POF, PM, AP, MEP, TEP, ADP_F, ADP_E), and some where the influence of leaching is major (HT_C) or even dominant (FEP, HT_{NC}, ET_{FW}). For brevity, in the following, these two groups of categories will be referred to as "Group T" and "Group L", respectively.

In both scenarios the transportation of C&DW and the avoided transportation of natural gravel are the most important processes for Group T categories and HT_C. In scenario a) they accounted for 60-95% of total impacts in absolute value. Crushing C&DW may in some cases contribute up to 30% of the total impacts, while capital goods only provide minor impacts, which suggests that from a life cycle point of view a system including several smaller crushing facilities may be recommended. Because leaching provides major impacts in Group L categories in scenario a), and especially for FEP, HT_{NC}, ET_{FW}, scenario b) provides lower (or at least comparable) impacts than a). Leaching of phosphate dominates eutrophication to freshwater bodies impacts (FEP); however, P transport is also limited by interactions with the soil, owing to

both adsorption and precipitation. Although several authors have studied phosphate interactions with soil (e.g. Matar et al., 1992; Tunesi et al., 1999) P retention in sub-soil has not yet been incorporated into fate modelling in the context of an LCA. Therefore, FEP impacts should be considered as overestimated. A value of PO_4 retention of 90% would result in scenarios a) and b) having comparable impacts (within the same order of magnitude).

Interestingly, impacts on marine and terrestrial eutrophication (MEP, TEP) are not affected by leaching at all, because impacts on MEP and TEP are only related to emissions of nitrogen compounds, and nitrogen was not measured in the leachates from C&DW. Therefore, our results for these categories are not as robust as for the other categories. Measurements of N in the leachates should be included in future studies, if eutrophication impacts are to be fully accounted for. Toxic leaching impacts were related mostly to oxyanions (As, V and Sb) rather than to metals. If the fate of Cr(VI) in the sub-soil was not taken into account based on Butera et al. (**III**)[3], Cr(VI) would represent the element responsible for the greatest impact in relation to both HT_C and ET_{FW}. It has been shown by Butera et al. (**I**) and (**II**) that C&DW may be very heterogeneous, especially in terms of release into water. The influence of this aspect was evaluated from an LCA perspective by adopting the 5^{th} and 95^{th} percentiles of the leaching data, instead of the average as in the reference scenarios. Percentage variations are reported in Table 6 (for the categories affected by leaching). The choice of leaching data results in variations in total impacts from 2 per cent to 70 per cent, i.e. in some cases it is non-negligible. However, the overall impacts did not change from environmental burdens to environmental savings, nor did they modify the relative contribution of the leaching process in the overall results. Other aspects might influence the overall outcome to a far greater extent; for example, ignoring the fate modelling of chromium (as discussed above) would result in an increase in HT_C and ET_{FW} impacts by 3400% and 43%, respectively, in scenario a), and 240% and

[3] This means that the reference modelling included that, of the total Cr released by C&DW, 99.7% is modelled as non-toxic Cr(III). If instead Cr had been inventoried based on direct measurements in the C&DW leachate as reported in Butera et al. (**II**) (i.e. 85% Cr(VI), then Cr(VI) would represent the element responsible for the greatest impact for both HT_C and ET_{FW}.

125% in scenario b). Variability of the leaching data appears as a factor of relatively lower importance.

Furthermore, if only toxic impacts actually taking place in the first 100 years were considered, i.e. the fraction "stored" in the sub-soil was ignored, the overall impacts for HT_C and ET_{FW} would decrease by 89% and 87%, respectively, in scenario a, while HT_{NC} would become an environmental saving. In scenario b the overall impacts of HT_C, HT_{NC} and ET_{FW} would decrease by 5%, 21% and 36%, which would affect the ranking of the two scenarios, since use in roads would now also rank better than landfilling for HT_{NC}.

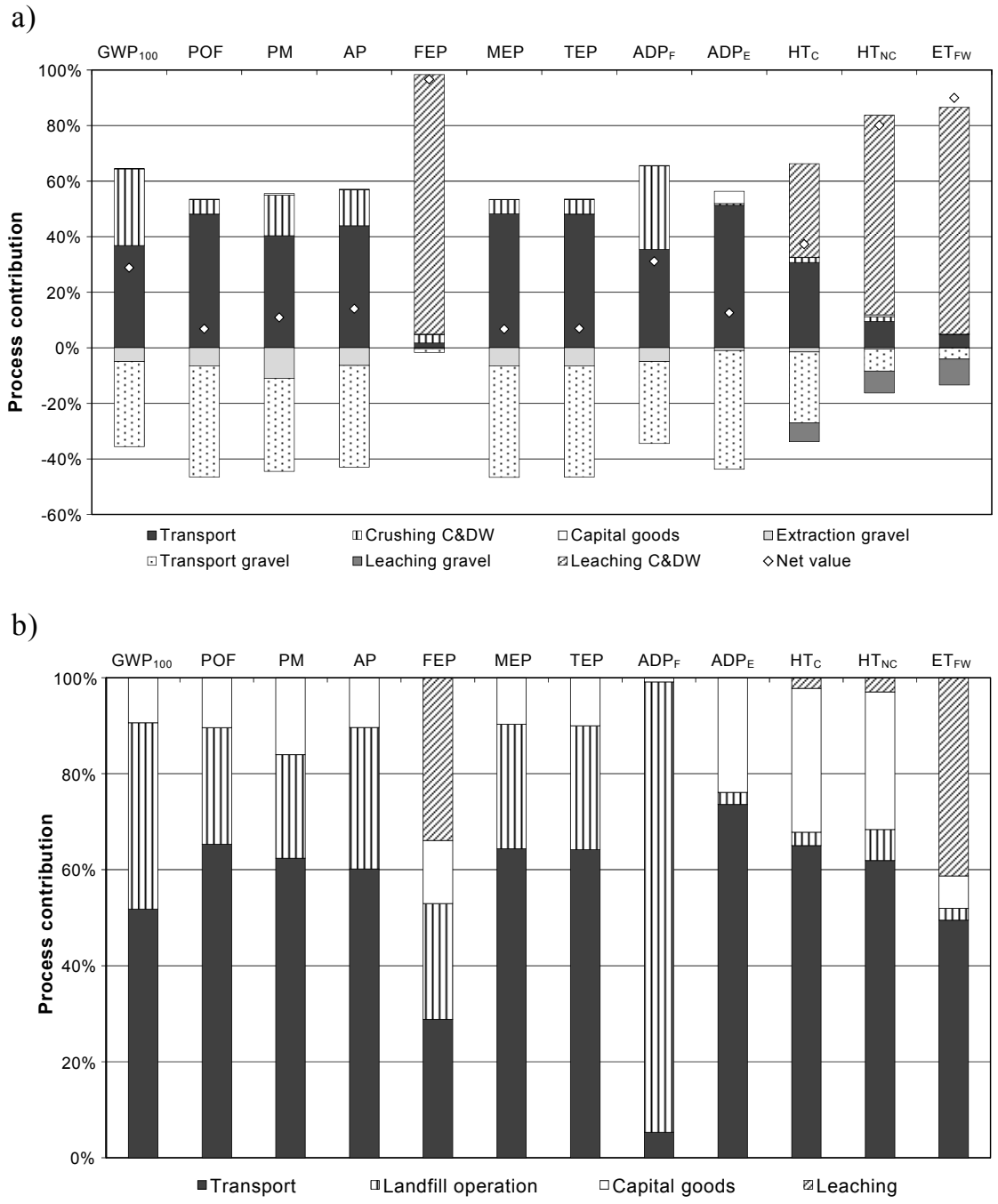


Figure 10 Environmental impact contribution of the different processes related to a) the utilisation of C&DW in road construction and b) the landfilling of C&DW (in percentages of the total impact). Only average leaching values are included. From Butera et al. (**IV**).

The carbonation of C&DW generally leads to increased oxyanions leaching (Mulugeta et al., 2011); in LCA terms, Butera et al. (**IV**) showed that HT_C and ET_{FW} impacts from C&DW utilisation might increase significantly as a consequence of carbonation. At the same time, however, GWP impacts would be reduced: while even a low degree of carbonation would result in negative

overall results, the extent of the GWP impact reduction is determined by the degree of carbonation, which in turn depends on storage conditions.

In terms of resource depletion (ADP_E, ADP_F), the utilisation of C&DW turned out to be a better option than its landfilling, with differences of one order of magnitude. However, the resources involved in this difference were mainly related to avoided transportation, rather than virgin aggregates. While the resource savings associated with the avoided production of virgin gravel and saved landfill space are evident based on common sense, these aspects are not captured by our LCA, as no characterisation factors for "virgin gravel" as a resource were developed in the resource depletion categories selected in Butera et al. (**IV**). Similarly to the case of water resources, global gravel resources might be sufficient, but the issue might be local scarcity, as these resources are not traded globally. LCIA methods typically refer to the continental or even the global scale (e.g. acidification, global warming), and thus they might not be fully appropriate for addressing local aspects such as scarcity of gravel. The ecological scarcity method (Frischknecht et al., 2009) addresses the environmental significance of virgin gravel savings based on gravel flows in Switzerland; however, as it reflects a very specific local situation, this method may not be applicable elsewhere. Care should therefore be taken when interpreting LCA results, by acknowledging what is included in the model and what has been left out.

Significance of LCA outcomes

Transportation has a significant impact on the results, and changing the distances assumed in the scenarios might significantly change the outcomes. For example, in cases where virgin aggregates are sourced from far away (further than 70 km), or in cases with short distances between a demolition site, a crushing plant and the road construction site (40 km instead of 60 km), recycling would provide environmental savings for Group T categories. On the other hand, distances greater than 150 km would make C&DW landfilling a better option for all categories except ADP_F. Similar conclusions would be obtained in cases where gravel has to be produced by excavating and crushing rocks, which would be a common scenario in countries without gravel pits. Therefore, the results concerning Group T categories – in terms of which scenario is better from an environmental standpoint – can easily be reverted by varying some of the model's most uncertain assumptions. On the other hand, Group L categories are not significantly affected by model assumptions

such as the choice of leaching data. This is partly related to the fact that Group T categories present both positive and negative impacts which are close to each other and may outbalance each other relatively easily. However, Group L categories may be influenced to a great extent by methodological aspects such as the environmental compartment to which emissions are inventoried, or the fate modelling of contaminants in one specific compartment. LCA practitioners should reflect on these aspects when dealing with systems involving long-term leaching. Using default values or default methods (for example, using the soil as the recipient for leaching emissions, just because they are physically directed to sub-soil) might not always be appropriate, and in some cases they might lead to serious mistakes.

6 Conclusions and recommendations

The goal of this PhD was to provide an assessment of potential environmental impacts related to C&DW utilisation. The main findings may be summarised as follows:

The large dataset of experimental data of composition and leaching of Danish C&DW collected herein revealed that:

- C&DW is characterised by significant variability, especially in terms of leaching. This may be related to the ageing level and also to the level of source segregation of the waste stream. On the one hand, proper assessment of this material should be based on large sampling campaigns. On the other hand, source segregation of C&DW should be ensured, as a clean concrete fraction would result in less leaching and potentially less impact on the environment.
- Leaching of selenium and, to a lesser extent, chromium, antimony, sulphate and chloride is critical for C&DW with respect to existing national and European regulations.
- PCBs are present even in recent C&DW, though not in critical concentrations. Their background levels seem to be non-negligible, proving that they spread to environmental matrices where they were not directly employed.

An investigation of percolation test methods focused on particle size and flow conditions revealed that standard up-flow columns are applicable when the focus is on cumulative release; however, estimations from standard up-flow percolation tests might be inaccurate for Ba, Mg, P, Pb and Zn. On the other hand, inaccurate quantification of early concentrations may be expected for Al, As, Ba, Cd, Cu, DOC, Mg, Mn, Ni, P, Pb, Sb, Se, Si and Zn when standard up-flow column tests are employed. Hence, in cases where accurate estimation of the concentrations is needed, testing conditions should be evaluated carefully.

Cr(VI) emitted by C&DW in road sub-bases is reduced in sub-soil, albeit with slow kinetics. Vertical migration of Cr(VI) in environmentally significant concentrations ($> 1\ \mu g \cdot L^{-1}$) is limited to the first 70 cm under the road sub-base, though in temperate climates, as well as in situations with high infiltration rates (storm events, unpaved roads, cracked asphalt cover), a migration of Cr(VI) by up to 2 m may be expected, which *might* represent a risk for

shallow groundwater bodies. The presence of an *actual* risk should be evaluated case-by-case based on local conditions such as the depth and flow of the groundwater, as well as its potential value as a drinking water resource.

From an LCA perspective, C&DW utilisation in road sub-bases does not typically provide net environmental benefits, in that gravel substitution does not compensate for the impacts of transportation, crushing and leaching. Nonetheless, C&DW utilisation is generally less damaging than landfilling, excluding toxicity impacts. The transportation of C&DW has significant impacts and should be minimised. In this respect, a C&DW management system, including several smaller crushing facilities, may be recommended, because environmental impacts from capital goods are minimal. On the other hand, leaching of oxyanion is responsible for toxicity to humans and freshwater species. Accurate modelling of Cr(VI) fate is essential for the correct estimation of LCA impacts. Carbonation of C&DW might further increase oxyanion leaching and at the same time decrease global warming impacts on environmental savings. The choice of which of the two aspects (toxicity or global warming impacts) to prioritise is a political choice for society. In LCA terms, leaching appears as the major impact from C&DW utilisation in roads, because it always results in a load on the environment, irrespective of the leaching data used. Even though the absolute values of leaching impacts might be uncertain, owing to the current methodology, toxic impacts are not greatly influenced by processes other than leaching (e.g. transportation, choice of marginal technology). On the other hand, energy-related impacts, such as global warming, for example, vary greatly depending on the transportation distances hypothesised or on the technology assumed for the marginal production of virgin aggregates. In other words, the importance of leaching is not sensitive to those parameters which in a *general* LCA study (as opposed to a case-specific study) are not known with certainty but can only be hypothesised (e.g. transportation distances, marginal technology, choice of leaching data). Only by improving the methodological aspects of the modelling (the environmental compartment to which emissions are inventoried, increased number of characterised inorganic substances) may the importance of leaching be estimated better. This highlights the need for methodological developments to model leaching impacts properly in an LCA. A final and important remark is that the LCA practitioner should be aware of the processes, substances and mechanisms included in their system boundaries as well as in the methods selected.

7 Perspectives

Based on the outcomes of this research, and on the experience acquired during this PhD, further research may be suggested on the following topics:

- Determining experimentally Cr's oxidation state at high pH values typical of mineral residues is not a trivial task. Most Cr speciation methods are pH-sensitive or based on cation-anion Cr(VI)-Cr(III) separation techniques, which are not applicable above pH 11, where Cr(III) might be present as negatively charged CrO_2^- or $Cr(OH)_4^-$. A development of *simple* analytical methods in this direction is therefore needed.
- During this project, Cr leaching was documented as quickly decreasing for L/S< 1 $L \cdot kg^{-1}$ TS and then stabilising around 10-50 $\mu g \cdot L^{-1}$ for L/S 2-14 $L \cdot kg^{-1}$ TS. While L/S of 14 $L \cdot kg^{-1}$ TS corresponds to approximately 100 years in a road sub-base scenario, and about 10 times more in a landfill scenario, longer time-frames might be relevant in some leaching studies. Other studies have shown that Cr leaching from MSWI air pollution control residues might increase for L/S> 50 $L \cdot kg^{-1}$ TS (Hykš et al., 2009b). Depending on the mineralogy, similar behaviour might be observed for C&DW and consequently needs to be investigated.
- The LCA study highlighted leaching as the aspect of greatest concern in C&DW utilisation; however, leaching modelling in LCA still presents some critical issues. The development of the LCA methodology is highly needed, especially concerning elemental coverage for inorganic pollutants and the number of environmental compartments included, in that USEtox CFs only exist for a few inorganic contaminants, and their fate and effects in environmental compartments such as sub-soil and groundwater have not been addressed yet.
- The oxidation state of oxyanions other than Cr was not investigated during this research, however some of them had a critical role in the LCA outcomes (especially As, Sb and V). One of the reasons was the lack of oxidation state-specific characterisation factors for other oxyanions, or incomplete coverage in USEtox. Further research in this direction is recommended.
- During this PhD, leaching from C&DW *as collected* was investigated, including partly carbonated samples. Pronounced differences were observed between fresher and partly aged C&DW for some elements, and at the

same time carbonation was found to affect significantly the carbon balance of cementitious materials. Given the research approach (investigate C&DW in conditions as close as possible to the subsequent utilisation) the extent of the carbonation level was not controlled, including, for example, progressively higher ageing levels. Thus, it was not possible to establish a precise relationship between the level of carbonation, CO_2 uptake and change in release for major, minor and trace elements, which was found ultimately to represent an important trade-off in LCA terms.

8 References

Allegrini, E., Butera, S., Kosson, D.S., van Zomeren, A., van der Sloot, H.A., Astrup, T.F., 2014. Life cycle assessment and residues leaching: importance of parameter, scenario and leaching data selection. Submitt. to Waste Manag.

Alonso-Santurde, R., Coz, A., Quijorna, N., Viguri, J.R., Andrés, A., 2010. Valorization of foundry sand in clay bricks at industrial scale. J. Ind. Ecol. 14, 217–230.

Anderson, R.A., 1997. Chromium as an essential nutrient for humans. Regul. Toxicol. Pharmacol. 26, S35–41.

Ball, J.W., McCleskey, R.B., 2003. A new cation-exchange method for accurate field speciation of hexavalent chromium. Talanta 61, 305–13.

Barceloux, D.G., 1999. Chromium. J. Toxicol. Clin. Toxicol. 37, 173–94.

Bartlett, R.J., James, B.R., 1979. Behavior of chromium in soils III. Oxidation. J. Environ. Qual. 8, 31–35.

Bartlett, R.J., Kimble, J.M., 1975. Behavior of chromium in soils I. Trivalent Forms. J. Environ. Qual. 5, 379–383.

Bary, B., Sellier, a., 2004. Coupled moisture—carbon dioxide–calcium transfer model for carbonation of concrete. Cem. Concr. Res. 34, 1859–1872.

Bhatty, J.I., 1995. Role of minor elements in cement manufacture and use. Res. Dev. Bull. RD109T, Portl. Cem. Assoc.

Birgisdóttir, H., 2005. Life cycle assessment model for road construction and use of residues from waste incineration. PhD Dissertation, Technical University of Denmark.

Birgisdóttir, H., Bhander, G., Hauschild, M.Z., Christensen, T.H., 2007. Life cycle assessment of disposal of residues from municipal solid waste incineration: recycling of bottom ash in road construction or landfilling in Denmark evaluated in the ROAD-RES model. Waste Manag. 27, S75–S84.

Bødker, J., 2006. Miljøvurdering af nye betontyper.

Bradl, H.B., 2004. Adsorption of heavy metal ions on soils and soils constituents. J. Colloid Interface Sci. 277, 1–18.

CEN, 2002. EN 12457-1 Characterisation of waste – Leaching – Compliance test for leaching of granular waste materials and sludges – Part 1.

CEN, 2004. EN 14997 Characterization of waste - Leaching behavior tests – Influence of pH on leaching with continuous pH-control.

CEN, 2014. DIN EN 14405:2014 Characterization of waste - Leaching behaviour test - Up-flow percolation test (under specified conditions).

Chandler, T.T., Eighmy, T.T., Hartlen, J., Hjelmar, O., Kosson, D.S., Sawell, S.E., van der Sloot, H.A., Vehlow, J., 1997. Municipal solid waste incinerator residues, Studies in. ed. Elsevier Science, Amsterdam, The Netherlands.

Clavreul, J., Baumeister, H., Christensen, T.H., Damgaard, A., 2014. An environmental assessment system for environmental technologies. Environ. Model. Softw. 60, 18–30.

Coelho, A., de Brito, J., 2012. Influence of construction and demolition waste management on the environmental impact of buildings. Waste Manag. 32, 532–541.

Danish EPA, 2011. Affaldsstatistik 2009 - Waste statistics 2009.

Danish EPA, 2013. Affaldsstatistik 2011 - Waste statistics 2011.

Danish Ministry of Environment, 2014. List of quality criteria in relation to contaminated soil and drinking water quality (Liste over kvalitetskriterier i relation til forurenet jord og kvalitetskriterier for drikkevand).

Delay, M., Lager, T., Schulz, H.D., Frimmel, F.H., 2007. Comparison of leaching tests to determine and quantify the release of inorganic contaminants in demolition waste. Waste Manag. 27, 248–55.

Dijkstra, J.J., Meeussen, J.C.L., Comans, R.N.J., 2009. Evaluation of a generic multisurface sorption model for inorganic soil contaminants. Environ. Sci. Technol. 43, 6196–6201.

Dijkstra, J.J., Meeussen, J.C.L., Van der Sloot, H., Comans, R.N.J., 2008. A consistent geochemical modelling approach for the leaching and reactive transport of major and trace elements in MSWI bottom ash. Appl. Geochemistry 23, 1544–1562.

Dutch Ministry of Housing Spatial Planning and the Environment, 2007. Soil Quality Decree (Besluit bodemkwaliteit).

Elovitz, M.S., Fish, W., 1994. Redox interactions of Cr(VI) and substituted phenols: kinetic investigation. Environ. Sci. Technol. 28, 2161–2169.

Engelsen, C., Lund, O., Breedveld, G., Mehus, J., Petkovic, G., Håøya, A., 2003. Leaching characteristic of unbound recycled aggregates - Preliminary study and ongoing research, in: Ortiz de Urbina, G., Goumans, J.J.J.M. (Eds.), WASCON 2003 San Sebastián, Spain. ISCOWA and Inasmet, pp. 177–186.

Engelsen, C., Mehus, J., Pade, C., Sæther, D., 2005. Carbon dioxide uptake in demolished and crushed concrete.

Engelsen, C., van der Sloot, H.A., Wibetoe, G., Justnes, H., Lund, W., Stoltenberg-Hansson, E., 2010. Leaching characterisation and geochemical modelling of minor and trace elements released from recycled concrete aggregates. Cem. Concr. Res. 40, 1639–1649.

Engelsen, C., van der Sloot, H.A., Wibetoe, G., Petkovic, G., Stoltenberg-Hansson, E., Lund, W., 2009. Release of major elements from recycled concrete aggregates and geochemical modelling. Cem. Concr. Res. 39, 446–459.

EU Commission, 2011. Supporting environmentally sound decisions for construction and demolition (C&D) waste management.

EU JRC, 2010. ILCD Handbook - General guide for Life Cycle Assessment - Detailed guidance, First edit. ed. Publications Office of the European Union.

EU JRC, 2011. ILCD handbook Recommendations for Life Cycle Impact Assessment in the European context - based on existing environmental impact assessment models and factors, First edit. ed. Publication Office of the European Union.

European Commission, 2002. Council Decision 2003/33/EC of 19 December 2002 establishing criteria and procedures for the acceptance of waste at landfills pursuant to Article 16 of and Annex II to Directive 1999/31/EC. EU.

European Commission, 2008. Directive 2008/98/EC of the European Parliament and of the Council of 19 November 2008 on Waste and repealing certain Directives, Hazardous Waste. EU.

Fendorf, S.E., 1995. Surface reactions of chromium in soils and waters. Geoderma 67, 55–71.

Fonseca, B., Teixeira, A., Figueiredo, H., Tavares, T., 2009. Modelling of the Cr(VI) transport in typical soils of the North of Portugal. J. Hazard. Mater. 167, 756–762.

Frischknecht, R., Steiner, R., Jungbluth, N., 2009. The Ecological Scarcity Method.

Fruergaard, T., Hykš, J., Astrup, T.F., 2010. Life-cycle assessment of selected management options for air pollution control residues from waste incineration. Sci. Total Environ. 408, 4672–4680.

Galvín, A.P., Ayuso, J., Agrela, F., Barbudo, A., Jiménez, J.R., 2013. Analysis of leaching procedures for environmental risk assessment of recycled aggregate use in unpaved roads. Constr. Build. Mater. 40, 1207–1214.

Galvín, A.P., Ayuso, J., García, I., Jiménez, J.R., Gutiérrez, F., 2014. The effect of compaction on the leaching and pollutant emission time of recycled aggregates from construction and demolition waste. J. Clean. Prod. 83, 294–304.

Galvín, A.P., Ayuso, J., Jiménez, J.R., Agrela, F., 2012. Comparison of batch leaching tests and influence of pH on the release of metals from construction and demolition wastes. Waste Manag. 32, 88–95.

Garrabrants, A.C., Sanchez, F., Gervais, C., Moszkowicz, P., Kosson, D.S., 2002. The effect of storage in an inert atmosphere on the release of inorganic constituents during intermittent wetting of a cement-based material. J. Hazard. Mater. 91, 159–185.

Garrabrants, A.C., Sanchez, F., Kosson, D.S., 2004. Changes in constituent equilibrium leaching and pore water characteristics of a Portland cement mortar as a result of carbonation. Waste Manag. 24, 19–36.

Goedkoop, M., Heijungs, R., Huijbregts, M., Schryver, A. De, Struijs, J., van Zelm, R., 2013. ReCiPe 2008 A life cycle impact assessment method which comprises harmonised category indicators at the midpoint and the endpoint level.

Goodwin, A.K., Chidiroglou, I., Laycock, E., O'Flaherty, F., 2008. Physical properties of demolition waste material. Proc. Inst. Civ. Eng. Constr. Mater. 161, 97–103.

Hansen, T.C., Lauritzen, E.K., 2004. Concrete Waste in a Global Perspective, in: Liu, T.C., Meyer, C. (Eds.), Recycling Concrete and Other Materials for Sustainable Development. American Concrete Institute, pp. 35–46.

Hauschild, M., Olsen, S.I., Hansen, E., Schmidt, A., 2008. Gone…but not away—addressing the problem of long-term impacts from landfills in LCA. Int. J. Life Cycle Assess. 13, 547–554.

Hauschild, M.Z., Goedkoop, M., Guinée, J., Heijungs, R., Huijbregts, M., Jolliet, O., Margni, M., Schryver, A., Humbert, S., Laurent, A., Sala, S., Pant, R., 2013. Identifying best existing practice for characterization modeling in life cycle impact assessment. Int. J. Life Cycle Assess. 18, 683–697.

Hellweg, S., Fischer, U., Hofstetter, T.B., Hungerbühler, K., 2005. Site-dependent fate assessment in LCA: transport of heavy metals in soil. J. Clean. Prod. 13, 341–361.

Hendriks, C.F., Janssen, G.M.T., 2001. Reuse of construction and demolition waste in the Netherlands for road constructions. Heron 46, 109–117.

Hillier, S.R., Sangha, C.M., Plunkett, B. a., Walden, P.J., 1999. Long-term leaching of toxic trace metals from Portland cement concrete. Cem. Concr. Res. 29, 515–521.

Hjelmar, O., 1990. Leachate from land disposal of coal fly ash. Waste Manag. Res. 8, 429–449.

Hjelmar, O., 2012. Description of the methodology used to set the leaching criteria for acceptance of waste at landfills for inert waste, landfills for non-hazardous waste accepting stable, non-reactive hazardous waste and landfills for hazardous waste listed in 2003/33/EC.

Hjelmar, O., Hykš, J., Wahlström, M., Laine-ylijoki, J., van Zomeren, A., Comans, R., Kalbe, U., Schoknecht, U., Krüger, O., Grathwohl, P., Wendel, T., Abdelghafour, M., Mehu, J., Schiopu, N., Lupsea, M., 2013. Robustness validation of TS-2 and TS-3 developed by CEN/TC351/WG1 to assess release from products to soil, surface water and groundwater.

Hyks, J., Astrup, T.F., Christensen, T., 2009a. Leaching from MSWI bottom ash: evaluation of non-equilibrium in column percolation experiments. Waste Manag. 29, 522–529.

Hyks, J., Astrup, T.F., Christensen, T., 2009b. Long-term leaching from MSWI air-pollution-control residues: leaching characterization and modeling. J. Hazard. Mater. 162, 80–91.

Jang, Y.C., Townsend, T.G., 2001. Occurrence of organic pollutants in recovered soil fines from construction and demolition waste. Waste Manag. 21, 703–15.

Jardine, P.M., Fendorf, S.E., Mayes, M. a., Larsen, I.L., Brooks, S.C., Bailey, W.B., 1999. Fate and transport of hexavalent chromium in undisturbed heterogeneous soil. Environ. Sci. Technol. 33, 2939–2944.

Kalbe, U., Berger, W., Eckardt, J., Simon, F.-G., 2008. Evaluation of leaching and extraction procedures for soil and waste. Waste Manag. 28, 1027–38.

Kalbe, U., Berger, W., Simon, F., Eckardt, J., Christoph, G., 2007. Results of interlaboratory comparisons of column percolation tests. J. Hazard. Mater. 148, 714–20.

Kayhanian, M., Vichare, A., Green, P.G., Harvey, J., 2009. Leachability of dissolved chromium in asphalt and concrete surfacing materials. J. Environ. Manage. 90, 3574–80.

Koletnik, D., Lukman, R., Krajnc, D., 2012. Environmental management of waste based on road construction materials. Environ. Res. Eng. Manag. 59, 42–46.

Kosson, D.S., van Der Sloot, H.A., Eighmy, T.T., 1996. An approach for estimation of contaminant release during utilization and disposal of municipal waste combustion residues. J. Hazard. Mater. 47, 43–75.

Kosson, D.S., van der Sloot, H.A., Garrabrants, A.C., Seignette, P.F.A.B., 2014. Leaching test relationships, laboratory-to-field comparisons and recommendations for leaching evaluation using the leaching environmental assessment framework (LEAF).

Krüger, O., Kalbe, U., Berger, W., Simon, F.-G., Meza, S.L., 2012. Leaching experiments on the release of heavy metals and PAH from soil and waste materials. J. Hazard. Mater. 207-208, 51–55.

Kucukvar, M., Egilmez, G., Tatari, O., 2014. Evaluating environmental impacts of alternative construction waste management approaches using supply-chain-linked life-cycle analysis. Waste Manag. Res. 32, 500–508.

Kumarathasan, P., McCarthy, G.J., Hassett, D.J., Pflughoeft-Hassett, D.F., 1990. Oxyanion substituted ettringites: synthesis and characterization; and their potential role In immobilization of As, B, Cr, Se and V. Mater. Res. Soc. Symp. Proc. 178, 83–104.

Kumpiene, J., Lagerkvist, A., Maurice, C., 2006. Retention of metals leached from municipal solid waste incineration (MSWI) bottom ashes in soils. Soil Sediment Contam. 15, 429–441.

Lagerblad, B., 2006. Carbon dioxide uptake during concrete life cycle – State of the art.

Laurent, A., Bakas, I., Clavreul, J., Bernstad, A., Niero, M., Gentil, E., Hauschild, M.Z., Christensen, T.H., 2014a. Review of LCA studies of solid waste management systems--part I: lessons learned and perspectives. Waste Manag. 34, 573–88.

Laurent, A., Clavreul, J., Bernstad, A., Bakas, I., Niero, M., Gentil, E., Christensen, T.H., Hauschild, M.Z., 2014b. Review of LCA studies of solid waste management systems--part II: methodological guidance for a better practice. Waste Manag. 34, 589–606.

Lindberg, R.D., Runnells, D.D., 1984. Ground water redox reactions: an analysis of equilibrium state applied to Eh measurements and geochemical modeling. Science (80-.). 225, 925–927.

Loijos, A., Santero, N., Ochsendorf, J., 2013. Life cycle climate impacts of the US concrete pavement network. Resour. Conserv. Recycl. 72, 76–83.

López Meza, S., Garrabrants, A.C., van der Sloot, H.A., Kosson, D.S., 2008. Comparison of the release of constituents from granular materials under batch and column testing. Waste Manag. 28, 1853–1867.

López Meza, S., Kalbe, U., Berger, W., Simon, F.-G., 2010. Effect of contact time on the release of contaminants from granular waste materials during column leaching experiments. Waste Manag. 30, 565–571.

López Meza, S., van Der Sloot, H.A., Kosson, D.S., 2009. The effects of intermittent unsaturated wetting on the release of constituents from construction demolition debris. Environ. Eng. Sci. 26, 463–469.

Manfredi, S., Christensen, T.H., 2009. Environmental assessment of solid waste landfilling technologies by means of LCA-modeling. Waste Manag. 29, 32–43.

Matar, A., Torrent, J., Ryan, J., 1992. Soil and fertilizer phosphorus and crop responses in the dryland mediterranean zone. Adv. Soil Sci. 18, 81–146.

Meijer, S.N., Ockenden, W. a, Sweetman, A., Breivik, K., Grimalt, J.O., Jones, K.C., 2003. Global distribution and budget of PCBs and HCB in background surface soils: implications for sources and environmental processes. Environ. Sci. Technol. 37, 667–672.

Meima, J. a., Comans, R.N.J., 1997. Geochemical modeling of weathering reactions in municipal solid waste incinerator bottom ash. Environ. Sci. Technol. 31, 1269–1276.

Meima, J.A., van Zomeren, A., Comans, R.N.J., 1999. Complexation of Cu with dissolved organic carbon in municipal solid waste incinerator bottom ash leachates. Environ. Sci. Technol. 33, 1424–1429.

Mercante, I.T., Bovea, M.D., Ibáñez-Forés, V., Arena, A.P., 2011. Life cycle assessment of construction and demolition waste management systems: a Spanish case study. Int. J. Life Cycle Assess. 17, 232–241.

Møller, J., Damgaard, A., Astrup, T.F., 2013. LCA af genbrug af mursten. Miljøprojekt 1512, 2013.

Monier, V., Hestin, M., Trarieux, M., Mimid, S., Domrose, L., Van Acoleyen, M., Hjerp, P., Mudgal, S., 2011. Service contract on management of construction and demolition waste.

Müllauer, W., Beddoe, R.E., Heinz, D., 2012. Effect of carbonation, chloride and external sulphates on the leaching behaviour of major and trace elements from concrete. Cem. Concr. Compos. 34, 618–626.

Mulugeta, M., Engelsen, C., Wibetoe, G., Lund, W., 2011. Charge-based fractionation of oxyanion-forming metals and metalloids leached from recycled concrete aggregates of different degrees of carbonation: a comparison of laboratory and field leaching tests. Waste Manag. 31, 253–258.

Mulugeta, M., Wibetoe, G., Engelsen, C., Lund, W., 2009. Fractionation analysis of oxyanion-forming metals and metalloids in leachates of cement-based materials using ion exchange solid phase extraction. Talanta 78, 736–742.

Nielsen, P., Kenis, C., Quaghebeur, M., Hermans, M., 2006. Environmental quality assessment of construction and demolition waste: comparison of percolation and batch

leaching tests, in: Ilic, M., Goumans, J.J.J.M., Miletic, S., Heynen, J.J.M., Senden, G.J. (Eds.), WASCON 2006 Belgrade, Serbia & Montenegro. ISCOWA, Belgrade.

Parkhurst, D.L., Appelo, C.A.J., 2013. Description of input and examples for PHREEQC Version 3 — A computer program for speciation, batch-reaction, one-dimensional transport, and inverse geochemical calculations, in: U.S. Geological Survey Techniques and Methods, Book 6, Chap. A43. U.S. Department of the Interior U.S. Geological Survey, p. 497.

Pivnenko, K., Eriksson, E., Astrup, T.F., 2014. Polychlorinated biphenyls (PCBs) in waste paper from Danish house-hold waste, in: 5th International Conference on Engineering for Waste and Biomass Valorisation, Rio de Janeiro, Brazil.

Pizzol, M., Christensen, P., Schmidt, J., Thomsen, M., 2011a. Impacts of "metals" on human health: a comparison between nine different methodologies for Life Cycle Impact Assessment (LCIA). J. Clean. Prod. 19, 646–656.

Pizzol, M., Christensen, P., Schmidt, J., Thomsen, M., 2011b. Eco-toxicological impact of "metals" on the aquatic and terrestrial ecosystem: A comparison between eight different methodologies for Life Cycle Impact Assessment (LCIA). J. Clean. Prod. 19, 687–698.

Rosenbaum, R.K., Bachmann, T.M., Gold, L.S., Huijbregts, M. a. J., Jolliet, O., Juraske, R., Koehler, A., Larsen, H.F., MacLeod, M., Margni, M., McKone, T.E., Payet, J., Schuhmacher, M., Meent, D., Hauschild, M.Z., 2008. USEtox—the UNEP-SETAC toxicity model: recommended characterisation factors for human toxicity and freshwater ecotoxicity in life cycle impact assessment. Int. J. Life Cycle Assess. 13, 532–546.

Sanchez, F., Garrabrants, A.C., Kosson, D.S., 2003. Effects of intermittent wetting on concentration profiles and release from a cement-based waste matrix. Environ. Eng. Sci. 20.

Sanchez, F., Gervais, C., Garrabrants, A.C., Barna, R.C., Kosson, D.S., 2002. Leaching of inorganic contaminants from cement-based waste materials as a result of carbonation during intermittent wetting. Waste Manag. 22, 249–60.

Saveyn, H., Eder, P., Garbarino, E., Muchova, L., Hjelmar, O., van der Sloot, H., Comans, R., van Zomeren, A., Hykš, J., Oberender, A., 2014. Study on methodological aspects regarding limit values for pollutants in aggregates in the context of the possible development of end-of-waste criteria under the EU Waste Framework Directive.

Schiopu, N., Tiruta-Barna, L., Jayr, E., Méhu, J., Moszkowicz, P., 2009. Modelling and simulation of concrete leaching under outdoor exposure conditions. Sci. Total Environ. 407, 1613–30.

Schör, H., 2011. Generation and treatment of waste in Europe 2008.

Schroeder, D.C., Lee, G.F., 1975. Potential transformations of chromium in natural waters. Water. Air. Soil Pollut. 4, 355–365.

Schwab, O., Bayer, P., Juraske, R., Verones, F., Hellweg, S., 2014. Beyond the material grave: Life Cycle Impact Assessment of leaching from secondary materials in road and earth constructions. Waste Manag.

Silva, R.V., de Brito, J., Dhir, R.K., 2014. Properties and composition of recycled aggregates from construction and demolition waste suitable for concrete production. Constr. Build. Mater. 65, 201–217.

Sleeswijk, A.W., Heijungs, R., Erler, S.T., 2003. Risk Assessment and Life-cycle Assessment: Fundamentally different yet reconcilable. Greener Manag. Int. 77–88.

Stripple, H., 2001. Life cycle assessment of road - A pilot study for inventory analysis. Göteborg.

Strufe, N., Trap, N., Lauritzen, E.K., 2006. Kortlægning af forurenende stoffer i bygge- og anlægsaffald.

Takeno, N., 2005. Atlas of Eh-pH diagrams - Intercomparison of thermodynamic databases.

Tam, V., Tam, C., 2006. A review on the viable technology for construction waste recycling. Resour. Conserv. Recycl. 47, 209–221.

Tiruta-Barna, L., Benetto, E., Perrodin, Y., 2007. Environmental impact and risk assessment of mineral wastes reuse strategies: Review and critical analysis of approaches and applications. Resour. Conserv. Recycl. 50, 351–379.

Trapp, S., Matthies, M., 1998. Chemodynamics and environmental modeling: an introduction. Springer, Berlin.

Tunesi, S., Poggi, V., Gessa, C., 1999. Phosphate adsorption and precipitation in calcareous soils: the role of calcium ions in solution and carbonate minerals. Nutr. Cycl. Agroecosystems 53, 219–227.

Van der Sloot, H.A., 1990. Leaching behaviour of waste and stabilized waste materials; characterization for environmental assessment purposes. Waste Manag. Res. 8, 215–228.

Van der Sloot, H.A., 2000. Comparison of the characteristic leaching behavior of cements using standard (EN 196-1) cement mortar and an assessment of their long-term environmental behavior in construction products during service life and recycling. Cem. Concr. Res. 30, 1079–1096.

Van der Sloot, H.A., 2002. Characterization of the leaching behaviour of concrete mortars and of cement-stabilized wastes with different waste loading for long term environmental assessment. Waste Manag. 22, 181–6.

Van der Sloot, H.A., Seignette, P.F.A.B., Meeussen, J.C.L., Hjelmar, O., Kosson, D.S., 2008a. A database, speciation modelling and decision support tool for soil, sludge, sediments, wastes and construction products: LeachXS^{TM}- Orchestra, in: Venice 2008 Second International Symposium on Energy from Biomass and Waste, Venice, Italy.

Van der Sloot, H.A., van Zomeren, A., Meeussen, J.C.L., Hoede, D., Rietra, R.P.J.J., Stenger, R., Lang, T., Schneider, M., Spanka, G., Stoltenberg-Hansson, E., Lerat, A., Dath, P., 2008b. Environmental CRIteria for CEMent based products.

Van Gerven, T., Van Baelen, D., Dutré, V., Vandecasteele, C., 2003. Influence of carbonation and carbonation methods on leaching of metals from mortars. Cem. Concr. Res. 34, 149–156.

Van Zomeren, A., Comans, R.N.J., 2004. Contribution of natural organic matter to copper leaching from municipal solid waste incinerator bottom ash. Environ. Sci. Technol. 38, 3927–3932.

Verschoor, A.J., Lijzen, J., Van den Broek, H.H., Cleven, R., Comans, R.N.J., Dijkstra, 2008. Revision of the Dutch Building Materials Decree: alternative emission limit values for inorganic components in granular building materials, in: 9th International Symposium on Environmental Geotechnology and Global Sustainable Development, Hong Kong, China.

Von Burg, R., Liu, D., 1993. Chromium and hexavalent chromium. J. Appl. Toxicol. 13, 225–230.

Wahlström, M., Laine-Ylijoki, J., Määttänen, A., Luotojärvi, T., Kivekäs, L., 2000. Environmental quality assurance system for use of crushed mineral demolition wastes in road constructions. Waste Manag. 20, 225–232.

Wehrer, M., Totsche, K.U., 2008. Effective rates of heavy metal release from alkaline wastes--quantified by column outflow experiments and inverse simulations. J. Contam. Hydrol. 101, 53–66.

Weng, C., Huang, C., Allen, H., Cheng, a, Sanders, P., 1994. Chromium leaching behavior in soil derived from chromite ore processing waste. Sci. Total Environ. 154, 71–86.

Wenzel, H., Hauschild, M.Z., Alting, L., 1997. Environmental Assessment of Products, Volume 1: Methodology, tools and case studies in product development. Chapman & Hall, London.

Wittbrodt, P.R., Palmer, C.D., 1996. Effect of temperature, ionic strength, background electrolytes, and Fe(III) on the reduction of hexavalent chromium by soil humic substances. Environ. Sci. Technol. 30, 2470–2477.

Zachara, J.M., Ainsworth, C.C., Cowan, C.E., Resch, C.T., 1989. Adsorption of chromate by subsurface soil horizons. Soil Sci. Soc. Am. J. 53, 418–428.

9 Papers

I **Butera S**, Christensen TH, Astrup TF. (2014). Composition and leaching of construction and demolition waste: inorganic elements and organic compounds. *Journal of Hazardous Materials*, 276, 302–311. doi:10.1016/j.jhazmat.2014.05.033

II **Butera S**, Hyks J, Christensen TH, Astrup TF. Construction and demolition waste: comparison of standard up-flow column and down-flow lysimeter leaching tests. *Submitted, 2014.*

III **Butera S**, Trapp S, Astrup TF, Christensen TH. Soil retention of hexavalent chromium released from construction and demolition waste in a road-base application scenario. *Submitted, 2014.*

IV **Butera S**, Christensen TH, Astrup TF. Life Cycle Assessment of construction and demolition waste management. *Submitted, 2014.*

In this online version of the thesis, the articles are not included but can be obtained from electronic article databases e.g. via www.orbit.dtu.dk or on request from:

DTU Environment
Technical University of Denmark
Miljøvej, Building 113
2800 Kgs. Lyngby
Denmark

reception@env.dtu.dk.

The Department of Environmental Engineering (DTU Environment) conducts science-based engineering research within four sections:
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The department dates back to 1865, when Ludvig August Colding, the founder of the department, gave the first lecture on sanitary engineering as response to the cholera epidemics in Copenhagen in the late 1800s.

DTU Environment
Department of Environmental Engineering
Technical University of Denmark

Miljoevej, building 113
2800 Kgs. Lyngby
Denmark

Phone: +45 4525 1600
Fax: +45 4593 2850
e-mail: reception@env.dtu.dk
www.env.dtu.dk